5.0 MASS BALANCE ACTIVITIES

5.1 ESTIMATED OVERALL MASS BALANCE OF RECYCLED U (INCLUDING Pu, Np, AND ^{99}Tc) BASED ON PREVIOUS ANALYSIS

Previous analysis by Egli, Smith[1], Bailey[2], and Parks[3] provide a good deal of perspective and insight regarding the flow and disposition of RU and the key constituents of interest (Pu, Np, ^{99}Tc) throughout the GDPs. Section 5.1 presents an analysis of the flow and disposition of these constituents at ORGDP based primarily on data and analysis presented in these referenced documents. This is provided for comparison with our current analysis presented in Section 5.2.

5.1.1 Uranium

Fig. 5.1-1 presents a summary of RU flow to and through ORGDP. Data are based on the Parks report issued as a preliminary draft in December 1999. The overall RU receipts at ORGDP was established to be 16,800 MTU. Approximately two-thirds of the UF_6 produced in the ORGDP feed plant was shipped to PGDP (including a small amount to PORTS), and only 5,350 MT of the RU converted to UF_6 in the ORGDP feed plant were fed to the ORGDP cascade.

Over its operating lifetime, 86,385 MTU of PGDP product were fed to the ORGDP cascade, along with 78,013 MTU of natural UF_6 feed, 41,947 MTU of other UF_6 (including re-feed), and the previously identified 5,350 MTU of RU. A portion of the 78,013 MTU of natural uranium feed was also processed in the ORGDP feed plant. This flow of natural uranium through the feed plant is not illustrated in Fig. 5-1, which is intended to highlight RU.

5.1.2 Plutonium

Fig. 5.1-2 presents the overall mass balance for Pu at ORGDP. A very high percentage of Pu is removed from RU in the feed plant and in the feed plant cylinder heels. In particular, the conversion from UF_4 to UF_6 results in the formation of nonvolatile compounds that result in the removal of most of the Pu, some of the Np, and a modest fraction of ^{99}Tc in the feed plant ash and in cylinder heels. Bailey estimates that 99.85% of the Pu is removed from RU in the feed plant at both PGDP and ORGDP. Smith and Parks estimate approximately 99.97% of the Pu is removed in the PGDP feed plant and/or retained in the UF_6 feed cylinders based on sampling of residue or "dust" in the PGDP cascade near feed points and analysis for Np and Pu. For

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[1] R. F. Smith, *Historical Impact of Reactor Tails on the Paducah Cascade*, KY/L-1239, Martin Marietta Energy Systems, Inc., Paducah Gaseous Diffusion Plant, March 1984.

[2] J. C. Bailey, "Radionuclides in the Equipment of the Oak Ridge Gaseous Diffusion Plant," Presentation to DOE Oak Ridge Operations and DOE Headquarters at the ORGDP, March 10, 1988.

[3] J. W. Parks, et al., Recycled Uranium Processed at the Department of Energy's Oak Ridge, Paducah, and Portsmouth Gaseous Diffusion Plants: Preliminary Report, U.S. DOE Oak Ridge Operations, December 1999.

perspective, the PORTS mass balance draft report[4] assumes 99.9% of the Pu is removed in the feed plant.

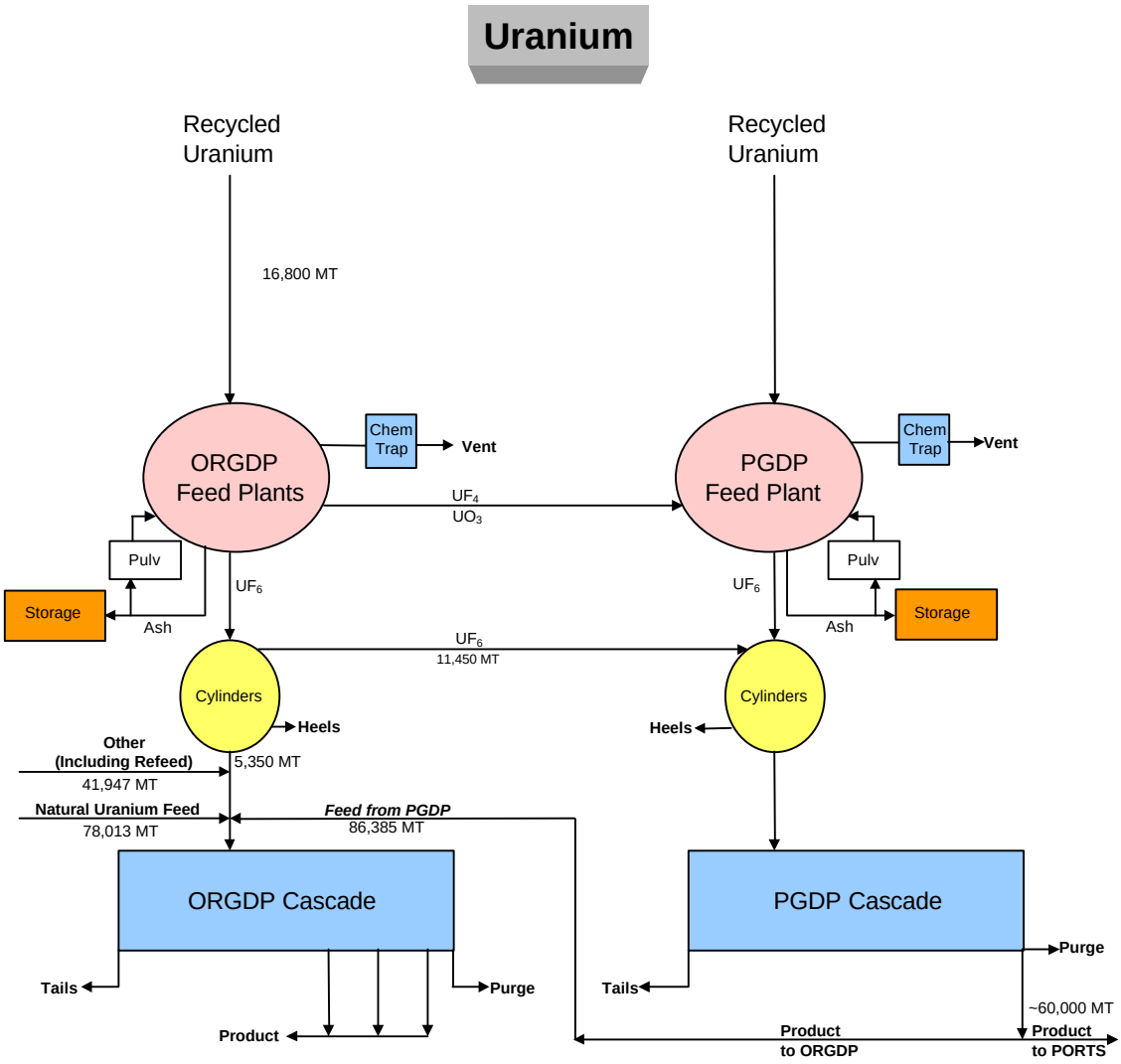
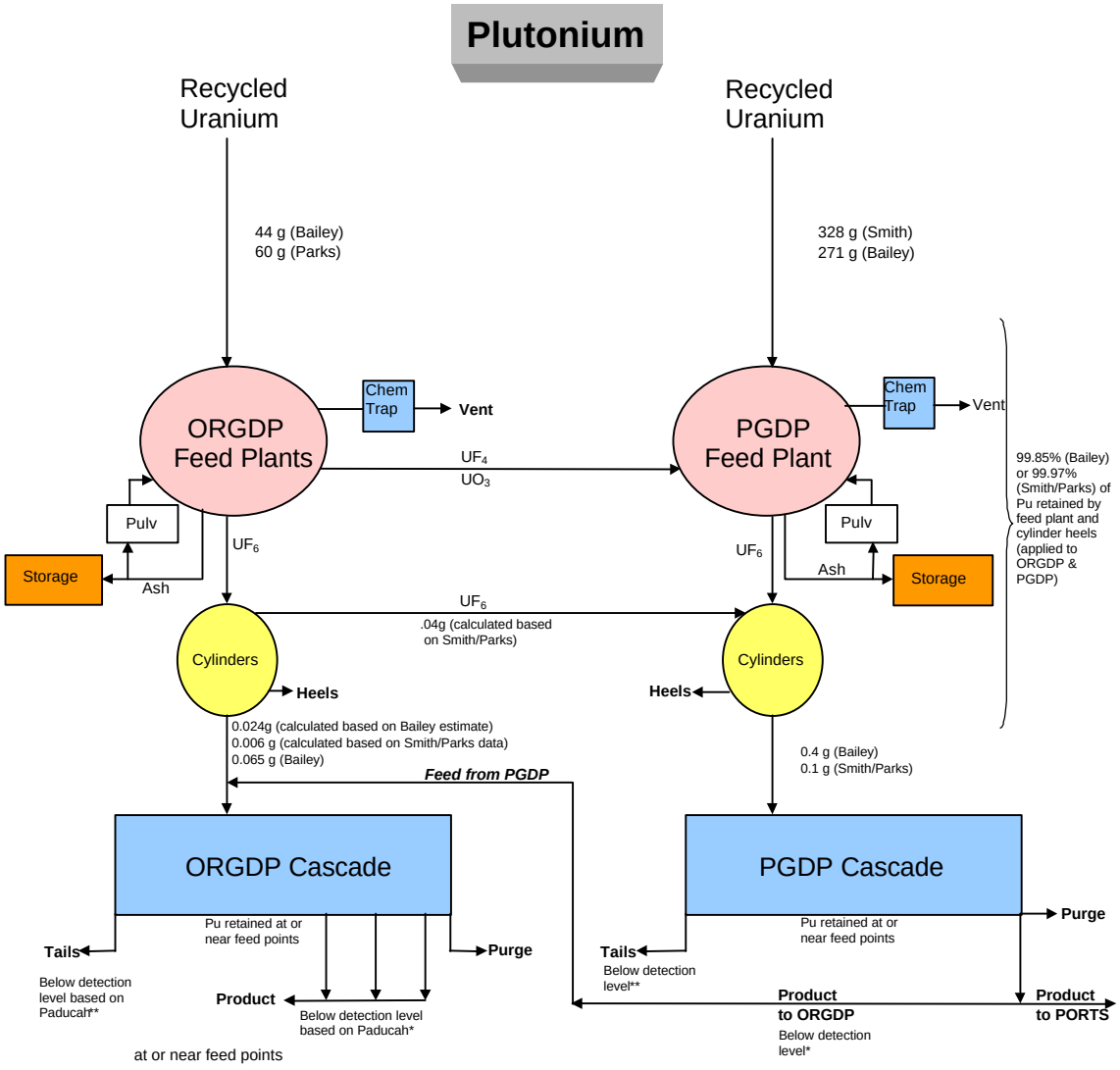


Fig. 5.1-1. Summary of Baseline RU Flow (Based on Parks Report).

[4] *Draft of Recycled Uranium Mass Balance Project Portsmouth, Ohio, Site Report*, BJC/PORTS-139, Bechtel Jacobs Company, LLC, Portsmouth Gaseous Diffusion Plant, March 2000



**Three product cylinders measured in 1973. Sixty product cylinders measured between 1975 - 1982; all were below detection level of 0.05 ppb initially and 0.01 ppb after 1980 except two cylinders, one showing 0.06 ppb and one showing 0.02 ppb (Smith).*

***Two tails cylinders measured in 1973 <.01 ppb Pu. Routine measurements since 1975 show <0.01 ppb Pu detection level (Smith).*

Fig. 5.1-2. ORGDP Baseline Mass Balance for Plutonium.

Extrapolating the Smith and Parks's analysis to ORGDP yields a projection of 0.006 g of Pu fed to the ORGDP cascade, versus the Bailey estimate of 0.065 g of Pu. Note that Bailey's estimate does not appear to take into account the fact that only about one-third of the RU received at ORGDP as oxide and converted to UF_6 was fed to the ORGDP cascade. Based on Bailey's estimate of 99.85% retained in the feed plant and 44 g received, we calculate 0.024 g fed to the ORGDP cascade.

All evidence indicates that essentially all Pu fed to the ORGDP and PGDP cascades was retained at or near the feed points. Based on analysis at PGDP, both product and tails samples indicate Pu concentrations are below detection levels. If trace quantities of Pu entered the cylinders it would tend to react and become fixed to the cylinder wall. Depending on the method of sampling, Pu on the cylinder wall might not be detected by sampling the contents.

5.1.3 Neptunium

Fig. 5.1-3 presents the overall mass balance for Np at ORGDP. The Bailey estimates for Np received at PGDP and ORGDP are somewhat lower than the Smith and Parks estimates. Bailey believed that documented information regarding the disposition of ^{99}Tc among the plants was probably the most reliable index for evaluating distribution of Pu and Np. Bailey used the ^{99}Tc values to calculate proportional amounts of Pu and Np. Smith estimated the Np concentrations in the RU based on limited available composite samples. These estimates appear to provide a reasonable bound on the range of Np receipts.

Both Bailey and Smith/Parks project that approximately 75% of the Np is retained in the feed plant and UF_6 feed cylinders and the remaining 25% is fed to the cascade. For perspective, the PORTS mass balance draft report makes the same assumption. Np fed to the cascade tended to plate out near the feed points. This is borne out by actual sample data from cascade equipment and is illustrated in the estimated Np distribution in the PGDP cascade shown in Fig. 5.1-4 (which is from Bailey).

No detectable levels of Np were found in the tails stream at PGDP based on 40 tails cylinders analyzed (Smith). Very minimum but detectable levels of Np were found in a few of the 60 product cylinders sampled at PGDP. Based on these results, it is assumed that an average of 2.5 ppb Np is included in the product stream from PGDP to ORGDP. This assumption is based on using one-half of the larger 5-ppb detection level available before 1980. This assumption results in projection of an additional 0.17 kg of Np fed to ORGDP.

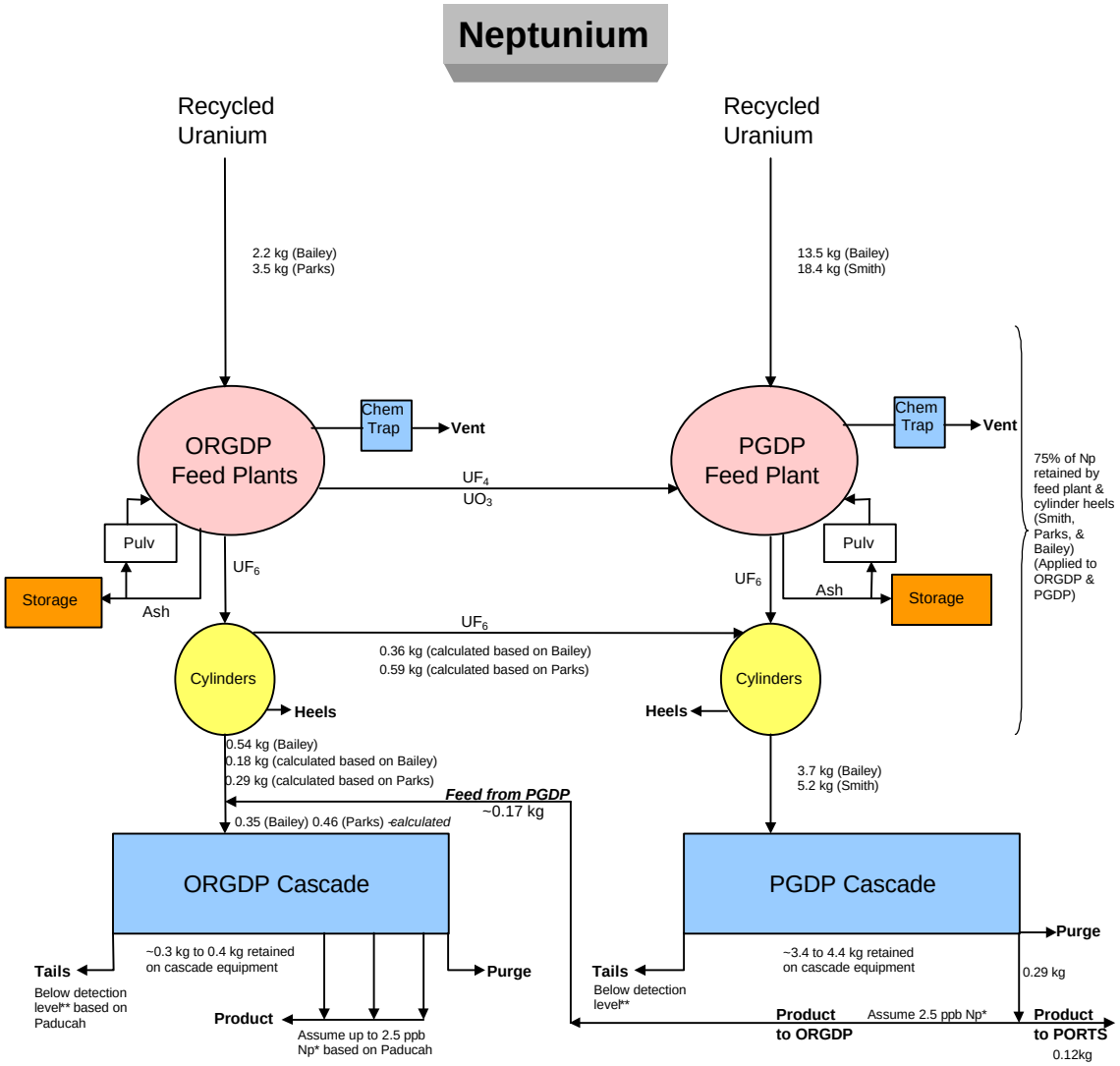
As illustrated in Fig. 5.1-4, Np fed to the ORGDP cascade is expected to have plated out quickly near the feed points. Based on an analysis of product cylinders at PGDP, it was estimated that less than 2.5 ppb Np might have progressed to the ORGDP product stream. Tails from the ORGDP cascade would be expected to be below the detection level for Np. If trace quantities of Np entered the cylinders, it would tend to react and become fixed to the cylinder wall. Depending on the method of sampling, Np on the cylinder wall might not be detected by sampling the contents.

5.1.4 Technetium

Fig. 5.1-5 presents the overall mass balance for ^{99}Tc at ORGDP. The projections of total ^{99}Tc in RU at PGDP are in good agreement at 670 kg (Bailey) and 661 kg (Smith and Parks). Approximately 15% of this is estimated by Smith to be retained in the feed plant and in cylinder

heels at PGDP. Bailey estimates similar retention of ^{99}Tc. For perspective, the PORTS mass balance draft report assumes that 10% of the ^{99}Tc is retained in the feed plant and cylinders.

For ORGDP, Bailey projects 86 kg of ^{99}Tc are received and fed to the cascade—which does not provide an allowance for retention in the feed plant and cylinder heels or for the portion of feed shipped to Paducah. However, after allowance for this retention and shipments to Paducah based on the Parks estimate, the estimated ^{99}Tc that was fed to the ORGDP cascade is 207 kg from Bailey and 152 kg estimated from Parks. Note that the total ^{99}Tc feed to the ORGDP cascade includes a significant contribution (121 kg) from ^{99}Tc contained in PGDP product shipped to ORGDP.



**60 product cylinders analyzed for Np at Paducah; a few exceeded 5 ppb detection level; highest measurement 27 ppb; most cylinders showed undetectable levels of Np; i.e., <1 and 5 ppb detection levels used (Smith). Assume average Np concentration was half of 5 ppb detection level.*

***40 tails cylinders analyzed for Np at Paducah; all were below 1 & 5 ppb detection levels (Smith).*

Fig. 5.1-3. ORGDP Baseline Mass Balance for Neptunium.

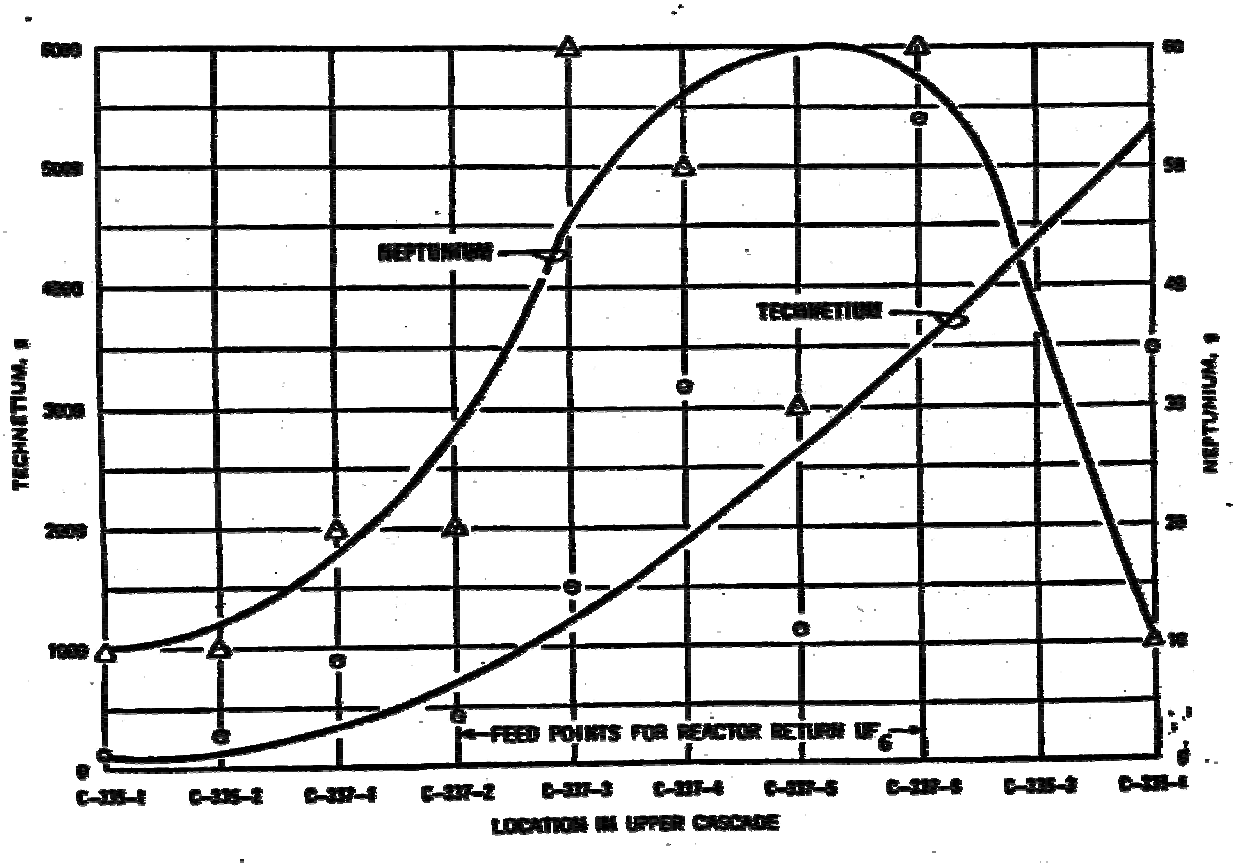
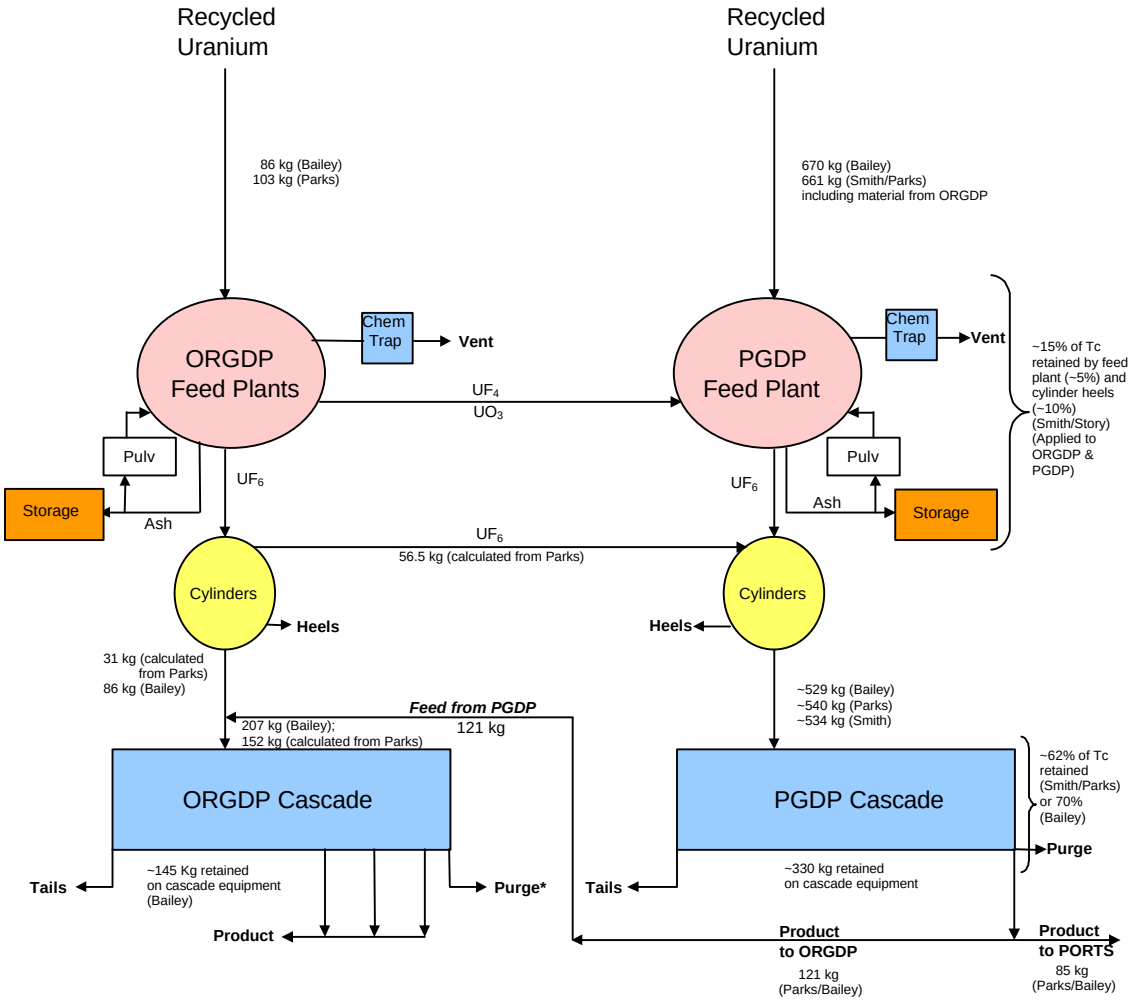


Fig. 5.1-4. Deposition of Neptunium and Technetium in the Upper Cascade of PGDP.

Technetium



**Most of the remaining Tc from the ORGDP cascade (~62 kg) is projected by Bailey to be in purge cascade equipment:*
~32 kg in K-311-1, K-310-3 system, and
~30 kg in K-402-8, K-402-9 or in the trapped material.
Some Tc was removed from the purge system by trapping.
Very little Tc is expected to have been included in the Product or Tails streams.

Fig. 5.1-5. ORGDP Baseline Mass Balance for Technetium.

Smith and Parks estimate that approximately 62% of the ^{99}Tc was retained on cascade equipment at PGDP, while Bailey estimates retention in the ORGDP cascade to be approximately 70% of the ^{99}Tc fed. The ^{99}Tc distribution in the cascade above the feed point at PGDP is also illustrated in Fig. 5.1-4.

The configuration of the ORGDP cascade is unique in that it includes an intermediate molecular weight gas purge system near the top of the cascade. Bailey projects that essentially all of the remaining 62 kg of ^{99}Tc at ORGDP accumulated in the purge cascade equipment or was trapped at the purge system.

5.2 ESTIMATED OVERALL MASS BALANCE OF RECYCLED URANIUM (INCLUDING Pu, Np, AND ^{99}Tc) BASED ON ANALYSIS OF BEST AVAILABLE DATA

Section 5.1 provided a baseline analysis developed from existing studies (primarily by Egli, Smith, Parks, and Bailey). The Section 5.2 analysis extends the evaluation by incorporating the best available data derived from a brief but intensive search of ORGDP records as previously described in Chapter 1.

This analysis presents a conservative estimate of the constituents contained in the RU considering both the previous analysis and new data that has been accumulated during this effort.

5.2.1 Uranium

As derived from the project team's investigation, the flow of RU to and through ORGDP is presented in Section 3.0 and summarized in Fig. 5.2-1. The total RU recognized to have been received at ORGDP has increased from the 16,800 MTU presented in the Parks report to 18,654 MTU. Most of the additional material was received in the form of UF_6 from foreign and other domestic sources.

The total RU fed to the ORGDP cascade is projected to be 5,915 MTU. It is important to note that approximately two-thirds of the RU received at ORGDP as oxide was shipped to PGDP as UF_6 after being processed in the ORGDP feed plant.

Table 5.2-1 presents the summary of RU received, shipped, and fed at ORGDP in tabular form.

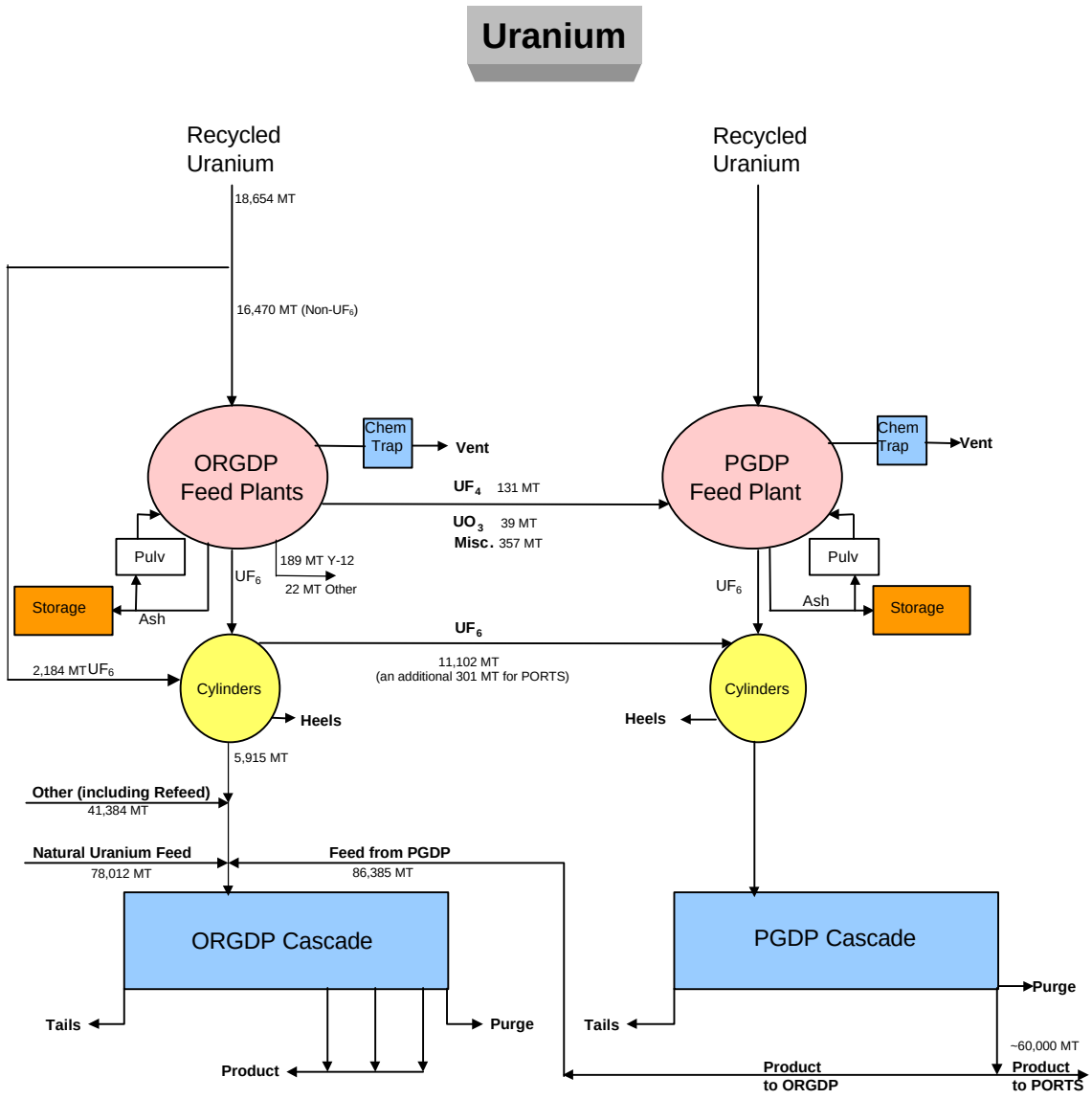


Fig. 5.2-1. Summary of Recycled Uranium Flow (Based on Section 3 and Table 5.2-1).

Table 5.2-1. Summary of RU Received, Shipped, and Fed at ORGDP Based on Analysis of Best Available Data

		MTU	
Receipts - Source Sites			
	Hanford (1952 - 58)	2,749	UO3(.646% - .666%)
	Hanford (1959 - 62)	1,527	UO3(.848% - .864%)
	Savannah River (1955 - 62)	10,290	UO3(.590% - .682%)
	Harshaw (1953 – 54)	1,702	UO3(.666% - .671%)
	Foreign (1969 – 88)	243	UF6 (~.65%)
		1,051	UF6 (Power Reactor Returns)
	Subtotal - Receipts Major Flows	**17,562**	
Receipts –Secondary Sites:			
	PGDP	98	UO3 (~.65%)
		88	UF4 (~.65%)
		887	UF6 (~.65%)
		11	Misc. (Includes Ash)
	PORTS	3	UF6 (~.65%)
	ORNL	3	UO3
		2	UF4
	Subtotal – Receipts Other Flows	**1,092**	
	Total Available	**18,654**	
Shipments To:			
	PGDP	39	UO3 (~.65%)
		122	UF4 (~.65%)
		9	UF4 (~.85%)
		10,577	UF6 (~.65%)
		39	UF6 (~.85%)
		486	UF6 (foreign returns)
		357	Misc (~.65%)
	PORTS	3	UO3 (~.65%)
		296	UF6 (~.65%)
		2	UF6 (~.85%)
	Y-12	189	Misc.
	Other	22	Misc.
	Subtotal - Shipments	**12,141**	
Feed to the Cascade		5,915	
	Total Available less Shipments and feed to Cascade	**598**	
	Inventory 3/31/1999	0	
	Cumulative losses and MUF	598	

5.2.2 Plutonium

Specific information on Pu in ORGDP RU receipts from Harshaw (October 1952 to December 1953), Hanford (February 1952 to June 1956), and Savannah River (December 1954 to July 1957) was obtained from correspondence from the ORGDP Laboratory Superintendent to the shipping site representative for analysis of RU as received. Based on these data, the weighted mean value of Pu concentration is 4.4 ppb in RU oxide shipped from these primary sources as shown on Table 5.2-2.

Table 5.2-2. Weighted Mean Value of Pu Concentration

Source	Average Pu *(ppb)*	Total Receipts *(kgs)*	Total Pu Received *(grams)*
Hanford	4.5	4,276,111	19.24
Harshaw	3.5	1,702,335	5.96
Savannah River	4.5	10,289,680	46.31
Weighted Average	4.4		

We should note that there is a discrepancy between Oak Ridge data and Hanford data for Pu content in RU from Hanford during the period of January to April 1953. The Oak Ridge data shows Pu concentrations during this period well above 10 ppb (see Figure 4.4-2). Hanford reported less than 10 ppb Pu concentration during this period.[5] If the Hanford data was used, the total projected Pu received at the ORGDP would have been closer to the Parks estimate of 60g.

The Oak Ridge data was used in this analysis. Other supporting evidence tends to confirm the higher estimate of Pu concentration from Hanford receipts during this period. For example, a January 6, 1953, memo discussing Pu buildup notes that "Pu content in RU from Hanford was less than 10 ppb except for approximately 30 tons which had 30 ppb."[6]

Other and foreign receipts were primarily in the form of UF_6 containing very low concentrations of Pu, Np, and fission products. The only specific data for these materials are identified by Smith, who reports the calendar year 1982 combined concentration of Np and Pu to average 6.1 ppb (<5 to 13) and states that for 1982 through 1983, the Pu ranged from <0.01 to 0.04 ppb and that Np ranged from <3 to 10 ppb. Based on this information, an average Pu concentration of 0.025 ppb and a Np concentration of 6.1 ppb have been projected for this material.

Table 5.2-3 presents the projected receipts of Pu per year. Based on this analysis, the total Pu receipts at ORGDP are projected to have been 71.5 g. (This estimate may be compared with the estimate of 44 g by Bailey and 60 g by Parks).

Essentially all of the Pu is believed to be retained in the feed plant and cylinder heels. The very low concentration of Pu in the foreign RU receipts (already converted to UF_6) provides support for this conclusion. The fraction of Pu retained in the feed plant and cylinder heels is estimated by Bailey at 99.85%; by the PORTS mass balance draft report at 99.9%; and by Parks at 99.97%. Based on these estimates, the Pu fed to the ORGDP cascade over the life of the plant is projected to be 0.01 to 0.04 g.

[5] "Reconciliation of Pu Data Between K-25 and Hanford," personal communication from Walt Scarbrough (Oak Ridge) to David Dodd (Hanford), 6/1/00.
[6] "Interim Report on Plutonium Buildup," memo from J.A. Marshall to A. P. Huber, January 6, 1953.

Table 5.2-3. Projected Annual Pu Received at ORGDP and Fed to the Cascade

Fiscal Year	Harshaw RU (kg)	Hanford RU (kg)	Savannah River RU (kg)	Other (PGDP, PORTS, ORNL, and Foreign) RU (kg)	Pu in Harshaw RU at 3.5 ppb (g)	Pu in Hanford RU at 4.5 ppb (g)	Pu in Savannah River RU at 4.5 ppb (g)	Pu in Other (PGDP, PORTS, ORNL, and Foreign) RU at 0.025 ppb (g)	Total Pu Received in RU at ORGDP (g)	Total Pu Fed to the ORGDP Cascade (g)
1952		99,970				0.45			0.45	
1953	1,402,761	578,249			4.91	2.60			7.51	
1954	299,574	1,115,345			1.05	5.02			6.07	
1955		526,475	271,949			2.37	1.22		3.59	
1956		323,882	2,538,844			1.46	11.42		12.88	
1957		98,218	2,635,163			0.44	11.86		12.30	
1958		7,201	1,077,065			0.03	4.85		4.88	
1959		261,253	828,250			1.18	3.73		4.91	
1960		609,775	1,677,456			2.74	7.55		10.29	
1961		611,020	1,121,645			2.75	5.05		7.80	
1962		44,722	139,308			0.20	0.63		0.83	
1963		1								
1964				Total Foreign						
1965				1969 - 1988						
1966				1,294,359				negligible	negligible	
1967				Total PGDP						
1968				and PORTS						
1969				1953 - 1970						
1970				1,086,962				negligible	negligible	
TOTALS*	**1,702,335**	**4,276,111**	**10,289,680**	**2,386,595**	**5.96**	**19.24**	**46.31**		**71.51**	**0.01 to 0.04**

** Numbers may not sum because of rounding.*

The estimates for Pu fed to the cascade are based on analysis of dust samples from Paducah cascade equipment confirming that plutonium alpha is approximately 1% of Np alpha.[7,8] Further analysis based on this information indicates that Pu fed to the cascade was ~0.011% of Np fed to the cascade, which led to the estimate of Pu retained in the feed plant and cylinder heels by Bailey, Parks, and the Portsmouth Mass Balance Report. Smith indicated this analytical approach resulted in a conservative upper limit of Pu fed to the cascade.

Recently, information relevant to these assumptions was requested from Gus Cook of Paducah. Cook advised that a classified Paducah document[9] was used to validate the (unclassified) Smith report data shown in Table 5.2-4.

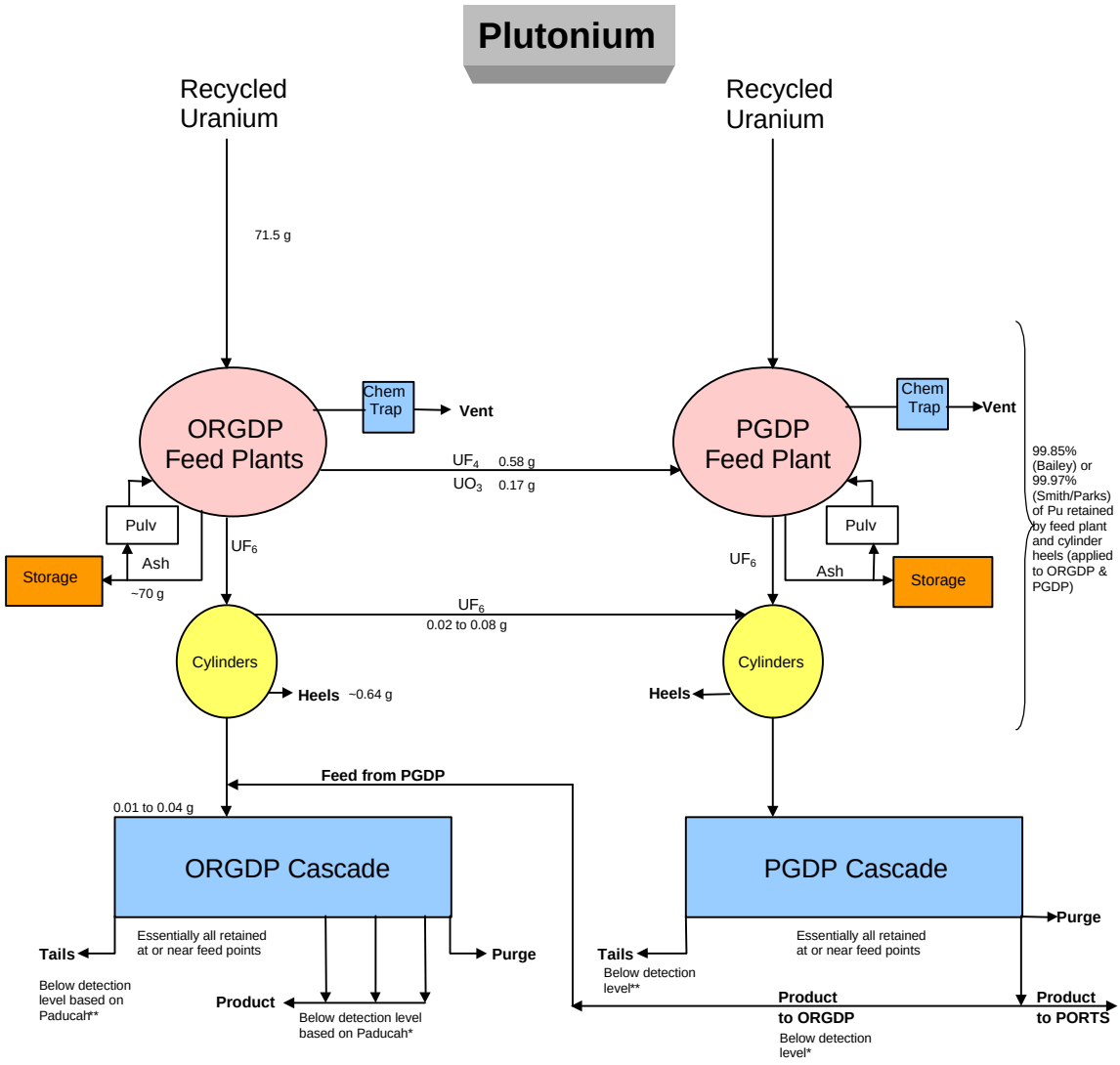
Table 5.2-4. Validated Data From Smith Report

	Remaining in Feed Plant Ash	Remaining in Cylinder Heels	Fed to the Cascade
Pu	99.0%	0.9%	0.1%
Np	25.0%	50.0%	25.0%
Tc	5.0%	10.0%	85.0%

The presence of Pu in ORGDP product and tails is expected to be well below detectable levels. Pu fed to the cascade is expected to have plated out on metal surfaces near the feed points and to have been removed with cascade equipment during the cascade improvement and cascade upgrade programs. This conclusion is supported by limited sample analysis of product and tails.

Fig. 5.2-2 presents the overall projected flow of Pu to and through ORGDP.

[7] Estimates of Transuranium Alpha Fed to the Paducah Cascade, memo from R. F. Smith to R. W. Levin, KY-L-411, April 19, 1966.
[8] Neptunium and Plutonium Plant Material Balance, memo from A. J. Lemonds to R. W. Levin, KY-L-565, July 19, 1971.
[9] Technetium and Plutonium Summary, KY-L-936 (classified), June 21, 1978.



**Three product cylinders measured in 1973. Sixty product cylinders measured between 1975 - 1982; all were below detection level of 0.05 ppb initially and 0.01 ppb after 1980 except two cylinders, one showing 0.06 ppb and one showing 0.02 ppb (Smith).*

***Two tails cylinders measured in 1973 <.01 ppb Pu. Routine measurements since 1975 show <0.01 ppb Pu detection level (Smith).*

Fig. 5.2-2. ORGDP Mass Balance for Plutonium.

5.2.3 Neptunium

The Smith report contains an Appendix 10 that summarizes the results of Np concentration analysis in RU measured subsequent to 1957. This summary is presented as Table 5.2-5.

Table 5.2-5. Neptunium Received in Reactor Tails and Fed to PGDP Cascade

Stream	Quantity of Reactor Tails *MTU*	Average Concentration *ppm Np, U Basis*	Concentration Range, *ppm Np, U Basis*	Apparent Np Received *(kg)*
Hanford and Savannah River FY 1957 to FY 1967	74,898	0.24	0.01 to 0.60	16.3
Hanford after FY 1967	22,326	0.09	0.05 to 0.27	1.8
Savannah River after FY 1967	1,890	0.12	<0.01 to 0.22	0.2
Enriched RU	2,154	0.05	0.01 to 0.11	0.1

During the early 1950s, ORNL Chemistry Division was exploring Np recovery from RU and from Purex Process Pu wastes. An ORNL paper prepared in 1957 by Lantz and Parker[10] provides information on four composite samples of Hanford UO_3 RU as shown in Table 5.2-6

Table 5.2-6. ORNL Analysis of Neptunium in Composite Hanford RU Samples

Sample Date	Sample Code	Description of Sample	Np^{237} ppm*
03/16/57	Composite 16 UA	Hanford UO_3	0.823
03/16/57	Composite 504	Hanford UO_3	0.869
05/03/57	Serial # 192-D	UO_3 Feed (Hanford)	1.098
06/18/57	UA-3615 21	UO_3 Feed (Hanford)	0.514

** Np concentration is presented in gram per ton in these historical reports, which we interpret to be grams per metric ton or parts per million.*

Lantz and Parker concluded that the theoretically calculated yield of Np in irradiated normal uranium is expected to be 2.5 to 3 gm of Np per kg of Pu. They assumed the level of Hanford metal to be 600 gm Pu per ton resulting in a Np yield of approximately 1.8 grams per ton; they concluded that almost one-half was being fed to the PGDP plant in RU.

Table 5.2-7 shows four additional data points on Np content from Savannah River calcined UO_3 (RU) at ORGDP from the ORNL Chemistry Division Semi Annual Progress Report for the period ending December 20, 1955, (ORNL-2046).

[10] P. M. Lantz and G. W. Parker, "Investigation of Paducah Ash and Metal Recovery Waste as a Large-Scale Source of Neptunium-237," Oak Ridge National Laboratory, July 1957.

Table 5.2-7. ORNL Analysis of Neptunium Content of Savannah River Calcined UO_3

Sample Date	Sample Code	Np^{237} ppm*
Approximately 1955 No specific date given but samples taken over several months prior to analysis	K-25, Lot 168	
	- Composite No. 1	0.694
	- Composite No. 2	0.682
	K-25, Lot 1196	
	- Trailer Load No. 1	0.747
	- Trailer Load No. 2	0.769

** Np concentration is presented in gram per ton in these historical reports, which we interpret to be grams per metric ton or parts per million.*

The report notes that the higher than expected Np concentration was not a transient condition as shown by the results from the composite samples, which were collected over several months.

Bailey asserts that documented information regarding the ^{99}Tc content in RU is probably the most reliable index for evaluating the distribution of Pu and Np. Bailey's analysis of Np received in RU oxide at ORGDP is 2.2 kg or approximately 0.13 ppm.

Finally, based on analysis of the French RU received at ORGDP in 1982 (Smith), Np received in foreign receipts in the form of UF_6 is projected to be approximately 6.1 ppb. Again, since this material is already converted to UF_6, it would be expected that most of the Pu and Np would have been removed from the UF_6 feed.

In summary, for the period when Hanford and Savannah River RU oxides were received at ORGDP, Smith projects a Np concentration of 0.24 ppm, Bailey calculated a Np concentration of 0.13 ppm, and the eight composite data points for Hanford and Savannah River RU in 1955-57 show an average Np concentration of 0.78 ppm. This is obviously a wide range of variation. The Smith estimates are based on monthly composite sample analysis for material received between 1957 and 1967. The much higher ORNL estimates are for eight composite samples performed in 1955 and March thru June 1957. The Bailey estimate does not appear to be supported by data.

This analysis is based on using the ORNL data as the representative Np concentration for 1952 through mid-1957 and the Smith data for mid-1957 through 1967. This approach is not meant to suggest that there was a dramatic reduction in Np concentration received in 1957, but rather that this is a conservative basis for estimating Np receipts based on data available.

Table 5.2-8 summarizes the projected receipts of Np at ORGDP on this basis. The total projected receipts based on this analysis are approximately 9 kg.

Table 5.2-8. Projected Np Received at ORGDP and Fed to the Cascade

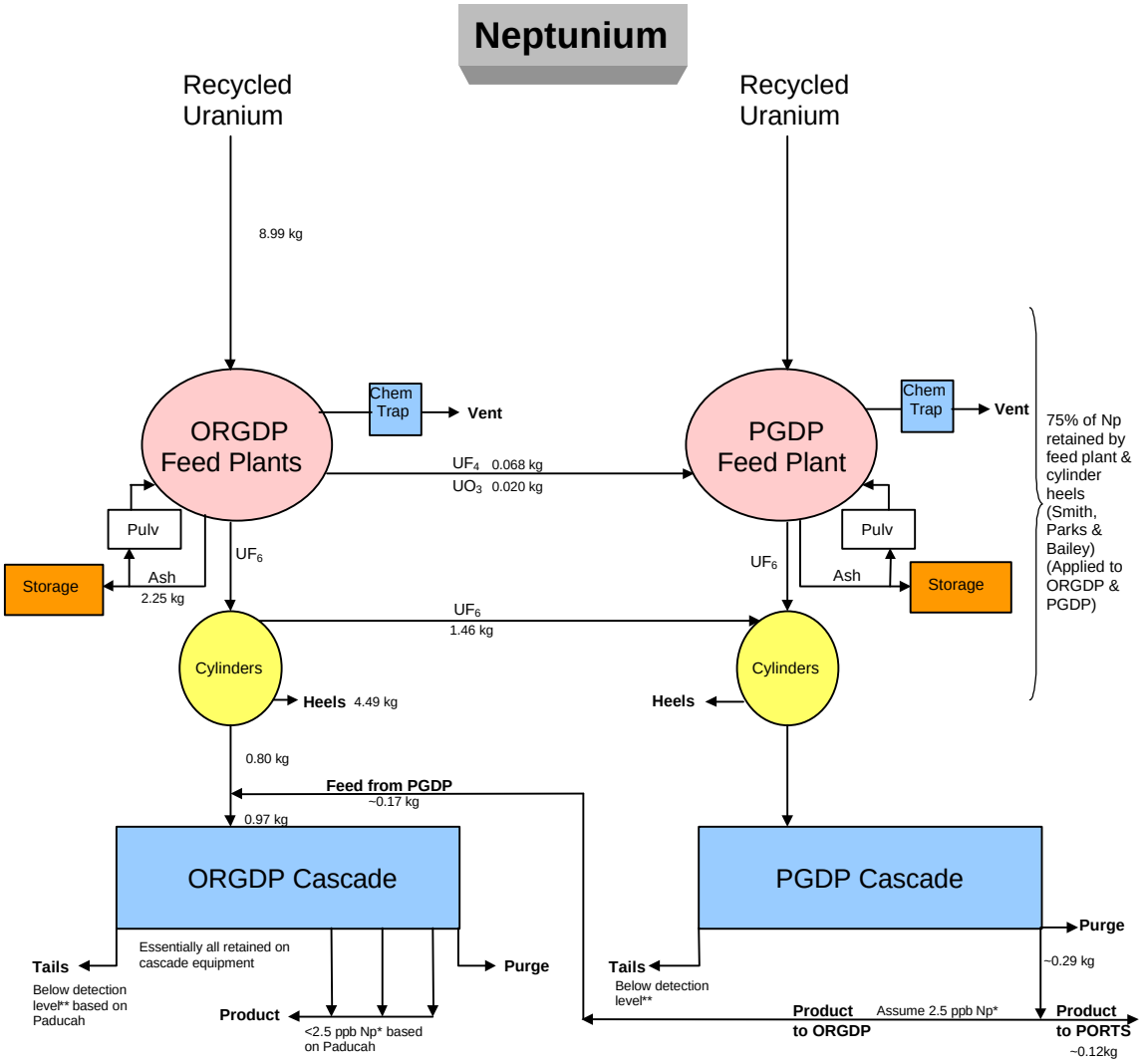
Fiscal Year	Harshaw RU *(kg)*	Hanford RU *(kg)*	Savannah River RU *(kg)*	Other (PGDP, PORTS, ORNL, and Foreign) RU *(kg)*	Estimated Np Concentration *(ppm)*	Np Received at ORGDP *(kg)*	Np Fed to ORGDP Cascade *(kg)*
1952		99,970			0.78	0.08	
1953	1,402,761	578,249			0.78	1.55	0.14
1954	299,574	1,115,345			0.78	1.10	0.10
1955		526,475	271,949		0.78	0.62	0.06
1956		323,882	2,538,844		0.78	2.23	0.20
1957		98,218	2,635,163		0.78 half yr. 0.24 half yr.	1.39	0.13
1958		7,201	1,077,065		0.24	0.26	0.02
1959		261,253	828,250		0.24	0.26	0.02
1960		609,775	1,677,456		0.24	0.55	0.05
1961		611,020	1,121,645		0.24	0.42	0.04
1962		44,722	139,308		0.24	0.04	
1963		1			0.24		
1964				Total Foreign			
1965				1969 - 1988			
1966				1,294,359	0.006	0.01	
1967				Total PGDP and			
1968				PORTS			
1969				1953 - 1970			
1970				1,086,962	0.006	0.48	0.04
TOTALS*	**1,702,335**	**4,276,111**	**10,289,680**	**2,386,595**		**8.99**	**0.80**

** Numbers may not sum because of rounding.*

Bailey and Smith/Parks project that 75% of the Np is retained in the feed plant and cylinder heels and that 25% is fed to the cascade. This estimate was based on (1) the quantity of neptunium fed to a group of feed cylinders during a closed cycle in which the cylinders were repeatedly filled with reactor tails UF_6, (2) a material balance across the feed plant for the period during which neptunium containing feed was processed, and (3) dust samples from various cascade equipment and cascade barrier. Smith estimated the range of Np retained in the feed plant and cylinder heels to be 60% to 90% and used an average of 75% in his analysis.

Based on this assumption, 0.97 kg (0.80 kg and 0.17 kg) of Np are projected to have been fed to the ORGDP cascade from RU received at ORGDP and from PGDP product as shown in Fig. 5.2-3. [Note that only about 36% of the RU oxide processed in the ORGDP feed plant is fed to the ORGDP cascade.] Fig. 5.2-3 presents the overall mass balance for Np at ORGDP based on this analysis, which is somewhat higher than the projections of Np fed to the ORGDP by Bailey (~0.36 kg) and Parks (~0.46 kg) as shown on Fig. 5.1-3.

Np fed to the ORGDP cascade is expected to have plated out quickly near the feed points as illustrated in Fig. 5.1-4. Based on analysis of product cylinders at PGDP, it was estimated that less than 2.5 ppb Np might have progressed to the ORGDP product stream. Tails from the ORGDP cascade would be expected to be below the detection level for Np.



**60 product cylinders analyzed for Np at Paducah; a few exceeded 5 ppb detection level; highest measurement 27 ppb; most cylinders showed undetectable levels of Np; i.e., <1 and 5 ppb detection levels used (Smith). Assume average Np concentration at Paducah was half of 5 ppb detection level.*

***40 tails cylinders analyzed for Np at Paducah; all were below 1 & 5 ppb detection levels (Smith).*

Fig. 5.2-3. ORGDP Mass Balance for Neptunium.

5.2.4 Technetium

5.2.4.1 Technetium Contained in PGDP Product Fed to ORGDP Cascade

The Smith/Parks estimate of 121 kg ^{99}Tc in the 86,385 MTU of PGDP product shipped to ORGDP is based on an estimated overall average ^{99}Tc content of approximately 1.4 ppm.

There are no data for 1953 through 1961, 1964 through 1971, and 1982 through 1985. ORGDP made ^{99}Tc measurements of PGDP product for the 5 months preceding the installation in January 1963 of a MgF_2 trap at PGDP to reduce the ^{99}Tc concentration in PGDP product. The average ^{99}Tc concentration in the PGDP product for the five months preceding the trap installation was 3.2 ppm. After installation of the trap, ORGDP continued measurements for 4 months during which the average ^{99}Tc concentration dropped to 0.15 ppm ^{99}Tc.

During the first 10.5 months of operation with the trap, 11.4 kg of ^{99}Tc was recovered from the trap. This rate of recovery implies that the ^{99}Tc concentration would have been approximately 4.7 ppm for the year without the trap. Assuming 3.2 ppm for 1 month and 0.15 ppm for 11 months, the estimated annual concentration of ^{99}Tc for 1963 was estimated to be 0.4 ppm.

Smith indicates that at some point not specified, attention to emptying the MgF_2 trap beds became lax, saturation resulted, and ^{99}Tc again increased in the product. Smith further indicates there was no RU fed to the PGDP cascade from 1965 to 1968 and in 1971.

Based on this information, an estimate of annual ^{99}Tc transferred to ORGDP in PGDP product was generated as shown on Table 5.2-9. The 3.2 ppm ^{99}Tc data from 1962 were attributed to PGDP product from 1953 through 1962 and 1969 through 1970. The ^{99}Tc concentration of 0.4 ppm in 1963 was previously explained. A modest increase to 1 ppm was assumed for 1964 through 1968 and for 1971 because, according to Smith, no RU was fed during 1965 through 1968 and in 1971. This estimate assumes a modest concentration during these years from residue in the cascade. Average ^{99}Tc concentrations presented for 1972 through 1982 were based on routine ^{99}Tc measurements made at PGDP as reported by Smith. For 1982 through 1985, ^{99}Tc concentrations were extrapolated from the data and trend for previous years.

The overall result, as shown in Table 5.2-9, is an estimate of 165 kg ^{99}Tc fed to the ORGDP cascade in PGDP product, an overall average concentration of 1.9 ppm. It is acknowledged that the previous estimate by Smith of 121 kg ^{99}Tc (1.4 ppm) may have been based on operational information that was available at PGDP—allowing a more precise estimate of ^{99}Tc concentration for years when data was not available. Lacking that additional insight, the project team judged the estimate presented in Table 5.2-9 to be reasonable.

Table 5.2-9. Projected Annual Tc Content in PGDP Product Fed to the ORGDP Cascade

Fiscal Year	PGDP Product Fed to ORGDP Cascade *(MTU)*	Average Tc *(ppm)*	Range of Tc *(ppm)*	Tc fed to ORGDP Cascade *(kg)*
1953	1,664	3.2	Assumption based on '62 data	5.30
1954	3,591	3.2	Assumption based on '62 data	11.50
1955	3,703	3.2	Assumption based on '62 data	11.80
1956	4,149	3.2	Assumption based on '62 data	13.30
1957	4,604	3.2	Assumption based on '62 data	14.70
1958	3,380	3.2	Assumption based on '62 data	10.80
1959	3,292	3.2	Assumption based on '62 data	10.50
1960	2,930	3.2	Assumption based on '62 data	9.40
1961	2,933	3.2	Assumption based on '62 data	9.40
1962	2,851	3.2	Average for last 5 months of '62 (Smith)	9.10
1963	2,871	0.4	MgF2 trap installed; 11.4 kg Tc trapped in 10.5 months; later dumping MgF2 trap became lax, saturation resulted, and Tc again increased in product (Smith)	1.10
1964	2,184	1.0	MgF2 trap in operation. Assumed 1.0 ppm for '64 and years when no recycled uranium fed	2.20
1965	2,126	1.0	No tails Fed (Smith)	2.10
1966	2,112	1.0	No tails Fed (Smith)	2.10
1967	1,931	1.0	No tails Fed (Smith)	1.90
1968	1,730	1.0	No tails Fed (Smith)	1.70
1969	2,713	3.2	Assumption based on '62 data	8.70
1970	2,637	3.2	Assumption based on '62 data	8.40
1971	2,832	1.0	No Tails Fed (Smith)	2.80
1972	2,782	0.67	0.19 to 1.7 (Smith)	1.80
1973	1,875	4.5	<0.1 to 20 (Smith)	8.40
1974	2,060	6.1	<0.2 to 20 (Smith)	12.60
1975	1,891	1.0	<0.2 to 3 (Smith)	1.90
1976	2,462	0.98	<0.3 to 3.7 (Smith)	2.30
1977	1,954	0.71	0.02 to 0.97 (Smith)	0.40
1978	1,131	0.19	<0.01 to 0.40 (Smith)	0.20
1979	1,218	0.14	<0.01 to 0.38 (Smith)	0.20
1980	2,099	0.09	<0.01 to 0.18 (Smith)	0.20
1981	4,945	0.01	<0.01 to 0.02 (Smith)	0.00
1982	3,457	<0.01		0.00
1983	2,689	<0.01		0.00
1984	1,294	<0.01		0.00
1985	2,295	<0.01		0.00
TOTALS*	**86,385**	**1.90**		**164.80**

** Numbers may not sum because of rounding.*

5.2.4.2 Technetium Received by ORGDP from Source Sites and Fed to ORGDP Cascade

The project team reviewed the Smith/Parks estimate of 103 kg ^{99}Tc in RU shipped to ORGDP based on revised projected receipts as shown on Table 5.2-10. Smith reports that hundreds of measurements were performed from 1959 to 1973 on ^{99}Tc content in depleted RU from Hanford and Savannah River and that essentially all of them clustered in a range of 4 to 10 ppm. Smith indicates the best ^{99}Tc concentration estimate for all depleted Hanford and Savannah River RU received through 1974 is 7 ppm ± 30%. Only five measurements were performed on enriched RU from Hanford with an average concentration of 16 ppm. These measurements provide the best estimate for that material.

`

Table 5.2-10. Projected Annual Tc Received by ORGDP Directly and Fed to the Cascade

Fiscal Year	Hanford, Savannah River, and Harshaw Depleted RU *(kg)*	Hanford Enriched RU *(kg)*	Other Receipts (PGDP, PORTS, ORNL, and Foreign) RU *(kg)*	Tc in Depleted RU at 7 ppm *(kg)*	Tc in Enriched RU at 16 ppm *(kg)*	Tc in Other Receipts of RU *(kg)*	Total Tc Received Directly at ORGDP *(kg)*	Tc from Receipts Fed to the ORGDP Cascade *15% of Tc received is projected to be retained in the Feed Plant & cylinder heels* *(kg)*
1952	99,970			0.7			0.7	0.2
1953	1,981,010		153,111	13.9			13.9	4.2
1954	1,414,919		21,396	9.9			9.9	3
1955	798,424		33,426	5.6			5.6	1.7
1956	2,862,726		30,389	20.0			20.0	6.1
1957	2,733,381		34,906	19.1			19.1	5.8
1958	1,084,266		29,020	7.6			7.6	2.3
1959	828,251	261,253	70,150	5.8	4.2		10.0	3.1
1960	1,677,456	609,775	2,091	11.7	9.8		21.5	6.6
1961	1,121,645	611,020	244	7.9	9.8		17.7	5.4
1962	139,308	44,722	10,511	1	0.7		1.7	0.5
1963	1		35,489					
1964			9,052					
1965			464					
1966								
1967								
1968								
1969 - 1988			1,951,071			6.7	6.7	5.7
TOTALS*	**16,268,126**		**2,386,595**	**103.2**	**24.5**	**6.7**	**134.4**	**44.6**

** Numbers may not sum because of rounding.*

RU received from other and foreign sources was primarily in the form of UF_6. The largest source of this material was from French Reactor Tails. The French receipts contained 0.041 ppm ^{99}Tc . Receipts from PGDP and PORTS are estimated at 6.6 kg and foreign receipts at 0.1 kg.

Based on these projected receipts and ^{99}Tc concentrations, the total ^{99}Tc in RU received directly at ORGDP was ~134.4 kg. Of this total, approximately 44.6 kg ^{99}Tc are expected to have been fed to the ORGDP cascade. [Note that only about 36% of the RU processed in the ORGDP feed plant is fed to the ORGDP cascade.]

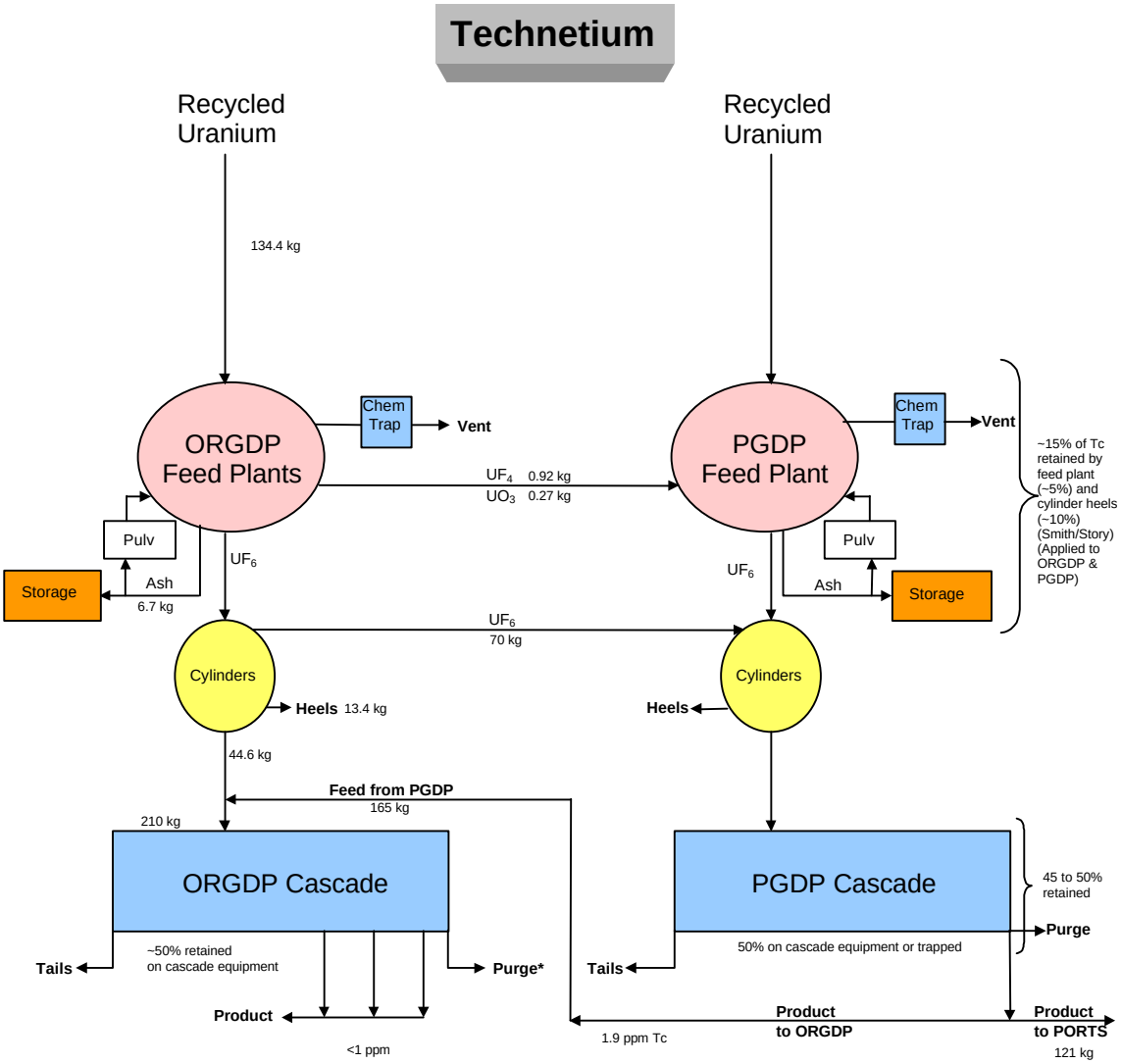
5.2.4.3 Overall Technetium Projection

Fig. 5.2-4 presents the overall projected flow of ^{99}Tc to and through ORGDP. It is projected that the overall flow of ^{99}Tc to ORGDP is approximately 134.4 kg in RU receipts of which 44.6 kg is estimated to have been fed to the ORGDP cascade, along with 165 kg received in PGDP product, for a total estimate of 210 kg of ^{99}Tc fed to the ORGDP cascade. An estimated 70 kg of ^{99}Tc were shipped to PGDP in 11,102 MT of UF_6. The total projected flow of ^{99}Tc to the ORGDP cascade of approximately 210 kg may be compared with the 207 kg projected by Bailey and 152 kg calculated from Parks data. [Note that Bailey in his analysis used the same estimate for ^{99}Tc received by ORGDP and that fed to the ORGDP cascade.]

It is important to note that the calculation of 165 kg of ^{99}Tc contained in PGDP product shipped to ORGDP, along with the equivalent estimate of 121 kg shipped to PORTS, results in a lower estimate of ^{99}Tc retained in the PGDP cascade (45 to 50%) than the projections of Smith/Parks (approximately 62%) or Bailey (approximately 70%), as shown in Fig. 5.1-5 in Section 5.1.

The most important concentration point for ^{99}Tc is in the ORGDP purge cascade equipment and the purge chemical trap. Other more modest projected concentration points include the feed plant, cylinder heels, and the ORGDP cascade equipment above the feed point.

Very little ^{99}Tc is expected to have been included in the product stream—certainly <1 ppm. In the tails stream, ^{99}Tc is expected to be below the detectable level.



**Most of the remaining Tc from the ORGDP cascade (up to ~110 kg) is projected to be in purge cascade equipment, i,e., the K-311-1, K-310-3 system and the K-402-8, K-402-9 system or in the trapped material. A significant quantity of Tc was removed from the purge system by trapping, but the specific quantity is not reported. Very little Tc is expected to have been included in the Product, certainly <1 ppm. Tc in the tails stream is expected to be below detectable levels.*

Fig. 5.2-4. ORGDP Mass Balance for Technetium.

5.3 POTENTIAL AREAS OF CONCENTRATION

Of the 18,654 MTU of RU received by ORGDP, the total estimated quantities of RU constituents of interest are:

- Pu: 71.5 g, based on data from RU receipts obtained from ORGDP Laboratory Superintendent Correspondence, of which only 0.01 to 0.04 g is estimated to have entered the cascade. The overwhelming majority of Pu was concentrated in the ash from the feed plant, and a small fraction was retained as cylinder heels. This estimate, based on ORGDP laboratory data, is modestly higher than the Parks estimate of 60 g.[11]

- Np: 9 kg, based on ORNL composite sample analysis prior to 1957 and Paducah sample analysis for 1957 to 1967, of which approximately 0.80 kg is estimated to have entered the ORGDP cascade, along with up to 0.17 kg of Np in PGDP product fed to the ORGDP cascade. Approximately 75% of the Np received as UO_3 is estimated to have remained in the feed plant ash and cylinder heels. Almost 1.5 kg of Np was shipped to PGDP in 11,102 MT of UF_6. Analysis for Np performed by ORNL in 1955 and early 1957 on composite samples of Hanford and Savannah River RU show much higher concentrations of Np (0.78 ppm Np average) than subsequent analysis reported by Smith (0.24 ppm Np average) for the period from mid-1957 through 1967.[12] This estimate is based on using the ORNL analysis for estimated Np concentration during 1952 through mid-1957 and the Smith analysis for the period from mid-1957 through 1963, when shipments from Hanford and Savannah River ceased.

- ^{99}Tc: 135 kg, based on measurements performed from 1959 to 1973 on Tc content in RU from Hanford and Savannah River, of which approximately 45 kg is estimated to have entered the ORGDP cascade in the RU feed stream—along with up to 165 kg of ^{99}Tc contained in PGDP product feed based on Paducah data for 1972 to 1982 and ORGDP measurements of ^{99}Tc in Paducah product during 1962 and 1963. Approximately 70 kg of ^{99}Tc was transferred to PGDP in 11,102 MT of UF_6. In the ORGDP cascade, ^{99}Tc tended to accumulate at the top of the cascade or to migrate to the purge cascade points at the high end of the plant configuration, where it was vented and trapped.

Throughout this analysis of constituents, the ORGDP Site Team has attempted to be conservative; i.e., to err on the side of over estimating rather than under estimating the concentration level of the constituents (Pu, Np, ^{99}Tc).

In performing the analysis, results are sometimes presented at a level of precision, based on the mathematics, which implies a greater level of confidence than the underlying data would justify. The Site Team has attempted to identify the underlying data used throughout the analysis so that the reader will have a basis for evaluating our results.

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[11] J. W. Parks, et al., *Recycled Uranium Processed at the Department of Energy's Oak Ridge, Paducah, and Portsmouth Gaseous Diffusion Plants: Preliminary Report*, U.S. DOE Oak Ridge Operations, December 1999.

[12] R. F. Smith, *Historical Impact of Reactor Tails on the Paducah Cascade*, KY/L-1239, Martin Marietta Energy Systems, Inc., Paducah Gaseous Diffusion Plant, March 1984.

Table 5.3-1 summarizes ORGDP receipts of RU, including constituents (i.e., Pu, Np, and ^{99}Tc) and Table 5.3-2 summarizes ORGDP shipments of RU, including constituents. Table 5.3-2 also summarizes the site accumulation, releases, and other unaccounted-for material.

Table 5.3-1 ORGDP Receipts of RU

Shipping Site	Material Form	Quantities Received (MTU)	Quantity of Pu (grams)	Quantity of Np (grams)	Quantity of Tc (grams)
Hanford	UO3	4,276	19.24	2,480	43,700
Savannah River	UO3	10,290	46.31	4,733	72,000
Harshaw	UO3	1,702	5.96	1,328	11,900
PGDP	UF6	887	trace	345	5300
	UO3	98	trace	51	700
	UF4	88	trace	46	600
	Misc	11	trace	6	100
PORTS	UF6	3	trace	1	trace
ORNL	UO3	3	trace	2	trace
	UF4	2	trace	1	trace
Foreign	UF6	1,294	trace	trace	50
TOTAL RECEIPTS		18,654	71.51	8,993	134,350

Table 5.3-2. ORGDP Shipments of RU

Receiving Site	Material Form	Quantities Shipped (MTU)	Quantity of Pu (grams)	Quantity of Np (grams)	Quantity of Tc (grams)
PGDP	UF6	11,102	0.05	1,460	70,000
	UF4	131	0.58	68	917
	UO3	39	0.17	20	273
	Ash	16	~70	2,250	6,380
	Misc	341	trace	trace	trace
PORTS	UF6	298	trace	trace	trace
	UO3	3	trace	trace	trace
Y-12	Misc	189	.48	102	840
ORNL	UO3	6	trace	trace	trace
	Misc	2	trace	trace	trace
Savannah River	UO3	11	trace	trace	trace
Fernald	UO3	2	trace	trace	trace
Foreign	UF6	1	trace	trace	trace
Feed To Cascade	UF6	5,915	0.03	800	44,600
TOTAL SHIPMENTS (and feed to the cascade)		18,056	~71.31	4,700	123,010
Site Accumulation, Releases, and other Unaccounted For		598	~0.20	4,293	11,340

NOTE: *PGDP Product in the amount of 86,385 MTU produced in part from RU was fed to the ORGDP cascade for further enrichment. This feed to ORGDP, which is in addition to the amounts in the above table, contained an estimated 170 grams Np and 165,000 grams Tc.*

ORGDP received a total of 18,654 MTU of RU. A total of 12,141 MTU has been identified as being shipped off-site. Included in the shipment total is 486 MTU in the form of UF_6 from foreign sources that remained at ORGDP until 1986, when the material was shipped to PGDP. A

total of 5,915 MTU has been identified as being fed to the plant cascade. Accountability records do not show any RU remaining in the ORGDP inventory. The remaining 598 MTU represents process losses and other unaccounted for RU. Process losses occurred that could have amounted to 1 to 2% of the RU processed. In addition, blending of RU with non-RU inventory (including both physical blending and the blending, or averaging, of assays of historical data in available documentation) resulted in a loss of the ability to identify RU inventories and transactions.

Based on historical data, it is estimated that nearly all of the incoming Pu (i.e., on the order of 99%) ended up in the fluorination tower ash while a small fraction was collected with the UF_6. Cumulative data also suggest that the bulk of Pu collected in the UF_6 feed cylinder remained in the UF_6 feed cylinder heel, allowing <0.1% of the incoming Pu to reach the ORGDP cascade. PuF_6 is easily reduced to non-volatile species. Consequently, any Pu entering the process with UF_6 was essentially immobilized at the feed points to the process. A total of 16 MTU of ash has been identified as being produced from RU. Historical records indicate that this ash was shipped off-site to Paducah, and most likely subsequently shipped to Fernald along with Paducah ash.

An interview with Mr. Joe Dykstra, who managed the feed plant during the period when RU was received and processed, revealed that the RU oxide was in general much less reactive than natural uranium oxide feed. As a result, the RU ash often contained a large fraction of unreacted UF_4. This ash was often pulverized and recycled to recover more of the uranium. One consequence of this poor reactivity would have been to reduce the concentration of ppb Pu/U in the ash.

Based on the projections previously noted by Smith[1] and Bailey[2], and confirmed by Cook[9], we estimate that over 70.7 g (approximately 99%) of Pu were retained in the feed plant ash. If all this Pu were contained in the 16 MTU identified as being shipped to Paducah, the concentration would be 4,400 ppb Pu/U. Analysis obtained from Fernald on ~22.5 MTU of ash shipped from Paducah to Fernald has an average batch concentration of 1300 ppb Pu/U and a maximum batch concentration of 7650 ppb Pu/U, which seems to bound the expected range.

The bulk of the incoming Np was also distributed between the tower ash (25%) and the cylinder heels (50%). In this case, however, a significant fraction of the Np (25%) was fed to the enrichment cascade (ORGDP or PGDP). A small increment of Np was also received in the PGDP product. Since NpF_6 is also easy to reduce compared to UF_6, Np was also retained on the high surface area barrier and other process surfaces near cascade feed points. Cascade data indicate that very small quantities of the Np likely reached the top of the enrichment cascade and were discharged to the environment. Very little Np is expected to have ended up in the UF_6 product. The estimated 4,513 g of site accumulation, releases, and unaccounted-for material is expected to have primarily accumulated in cylinder heels. Washing of the cylinder heels would have resulted in release of this material in sludge. However, historical records do not indicate that the UF_6 feed cylinders were washed at ORGDP.

Approximately 85% of the ^{99}Tc received in the RU was fed to the cascade (ORGDP or PGDP). ^{99}Tc formed volatile and semi-volatile chemical compounds that tended to migrate to the top of the enrichment cascade with the enriched uranium. Consequently, a larger fraction of ^{99}Tc was discharged to the environment. Chemical traps installed at the top of the enrichment cascade in the 1960s retained up to 80% of the ^{99}Tc in the process vent gas. The estimate of 11,340 g of site accumulation, releases, and other unaccounted-for material would include losses in cylinder heels. Washing of the cylinder heels would have resulted in release of this material in sludge.

From the feed points, ^{99}Tc migrated primarily toward the top of the cascade. Overall, approximately 50%of the ^{99}Tc fed to the ORGDP cascade is expected to have been plated out throughout the upper cascade as shown in Fig. 5.3-2.

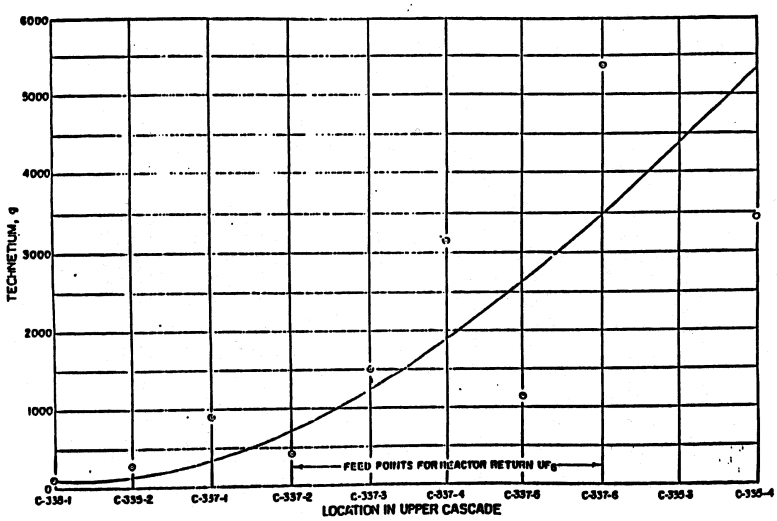

Fig. 5.3-2. Deposition of Tc in the Upper Cascade of PGDP.

Because of the unique configuration of the ORGDP cascade, the primary concentration of ^{99}Tc is expected to have occurred in the purge cascade equipment at the top of the enrichment plant. Specifically, the K-311-1 and K-310-3 systems and the K-402-8 and K-402-9 systems are likely concentration points. Portions of the ^{99}Tc were trapped at the purge cascade (approximately 100 pounds of ^{99}Tc and uranium compounds were trapped, but the fraction of this material which is ^{99}Tc is not known).

No detectable levels of ^{99}Tc are expected to have been contained in the tails. It is reasonable to assume that some ^{99}Tc was present in the product because of evidence of ^{99}Tc deposited throughout the upper cascade. However, unlike PGDP, which produced product containing an average of 1.9 ppm ^{99}Tc, at ORGDP the purge system, rather than the product withdrawal point, was the primary ^{99}Tc concentration point. As a result, very little ^{99}Tc is expected to have been in the product stream at ORGDP—certainly <1 ppm.

5.4 POTENTIAL FOR WORKER EXPOSURE

5.4.1 Film Badge Exposure

To gain insight into the numbers of workers who could potentially have been exposed to RU constituents at ORGDP, the project team reviewed *ORGDP Quarterly Reports* from FY 1952 to FY 1963.[13] The reports provided quarter-ending total ORGDP employment numbers, which are shown in Figure 5.4-1.

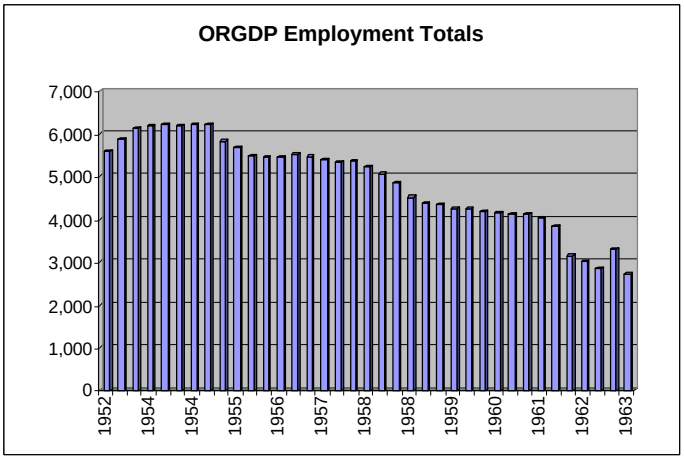

Fig. 5.4-1. ORGDP Employment Totals.

The *Quarterly Reports* also provided information generally documenting the number of personnel exposures above the Plant Allowable Limit (PAL) and/or providing exposure summaries relative to other points in time (i.e., an increase or decrease from a previous reporting period). Table 5.4-1 summarizes discussions on personnel exposures in the *Quarterly Reports*.

[13] *ORGDP Quarterly Reports*, 1952 to 1963

Table 5.4-1. Personnel Exposures and Explanations Extracted from *ORGDP Quarterly Reports*

Fiscal Year	Quarter	Exposures	Exposure Explanation
1954	Q1		There continues to be no clinical evidence of permanent damage to personnel from exposure to atmospheric contaminants.
	Q2	15	15 film badge exposures above PAL occurred.
	Q3		Decreases in personnel exposure to penetrating radiation were noted as were reductions in the levels of both penetrating radiation and alpha contamination.
	Q4		Increase in plant penetrating rad levels and the number of over-PAL rad exposures, but average exposure for plant personnel decreased for second consecutive quarter. Alpha contamination also showed slight decrease.
1955	Q1		Penetrating rad levels showed a considerable decrease; Reductions were noted in the number of spot air samples which indicated air-borne uranium activity in excess of the PAL.
	Q2		Increase in the number of personnel exposures to penetrating radiation levels slightly in excess of the PAL; the average exposure of plant personnel of 8 mrep per week was ~ 1/2 the corresponding figure for 1953.
	Q3		plant rad and contamination indices and personnel exposures to penetrating rad decreased to the lowest values since 1952.
	Q4	1	1 exposure in excess of PAL; decon operations resulted in continued decreases in rad surface contamination, penetrating rad levels, and airborne radioactivity; average film badge exposure decreased to only 2 mrep as compared to an average of 7.8 mrep for 1954.
1956	Q1		Average film badge exposure decreased to 0.4 mrep/week.
	Q2	7	7 personnel exposures to penetrating radiation in excess of PAL brought the year's total to 9. The max individual indicated exposure of 7.8 rep, although the highest in several years, is not considered indicative of injury to the employee involved.
	Q3	10	10 personnel exposures to penetrating rad in excess of the PAL.
	Q4	6	Average personnel exposure to penetrating radiation remained unchanged; 6 exposures in excess of PAL.
1957	Q1	12	None
	Q2	0	Although penetrating rad levels have exhibited a general upward trend since 6/55 (as a result of normal accumulation of U daughter products in K-1131 feed plant), the average film badge exposure for the year was only slightly above that for 1955.
	Q3		The utilization in the K-1131 feed plant of reprocessed fuel materials having both unfavorable chemical properties and an unusually high content of U-238 daughter products, resulted in increased maintenance activities and in somewhat higher rad fields than normal. This is reflected by increases in the penetrating rad index, the average film badge exposure to penetrating rad, and the number of exposures in excess of the PAL, and in the number of positive urinary analyses for U and fluorides. However, none of the exposures noted were indicative of injury to employees.
1958	Q2	11	Radiation levels associated with normal accumulation of uranium daughter products in the K-1131 Plant increased during 1957. Personnel exposures to penetrating radiation in excess of plant acceptable limit. This was a decrease from 17 similar incidents in 1956.

Of particular interest is an entry for the second quarter of 1956 that documents an exposure of 7.8 rad equivalent person (rep) as the highest such exposure in "several years." Three other discussions of interest are noted in the second and third quarters of 1957 and in the second quarter of 1958. All three of these entries mention exposures relative to ^{238}U daughter-product accumulations from the use of reprocessed fuel materials at the K-1131 Feed Plant.

5.4.2 Urinalysis Data

Results of urinalyses for uranium, alpha activity, neptunium, plutonium and technetium are listed in an ORGDP historical database for the period of 1948 to 1992 providing evidence of a significant, established personnel monitoring program in the years that RU was processed. Throughout the plant history the primary tools (Table 5.4-2) for monitoring employee exposures appear to have been analysis for total alpha and analysis for uranium. Limited data is available

from plutonium and neptunium analyses, which may have been performed only in cases in which specific exposure was suspected. Results for technetium represent a large population, however available data was limited to years FY 1978 to FY 1993.

Table 5.4-2. ORGDP Urinalysis Testing – 53 Departments.

Analysis	Period for Which Data Exists	Average Number of Employees Tested Per Year	Average Number of Tests Per Year
alpha activity	1948 - 1993	965	3,548
uranium	1948 – 1991	995	3,618
plutonium	1954 - 1963	5	14
neptunium	1963	32	39
technetium	1978 - 1993	756	4369

Of the persons tested for alpha activity over 90% were also tested for uranium. The number of tests performed annually and also the number of persons tested at ORGDP varied over the time period from CY 1948 to FY 1992. Figures 5.4-2 depicts the number of tests performed for alpha activity and the number of personnel tested. Figure 5.4-3 shows the number of tests performed for uranium during the same time period.

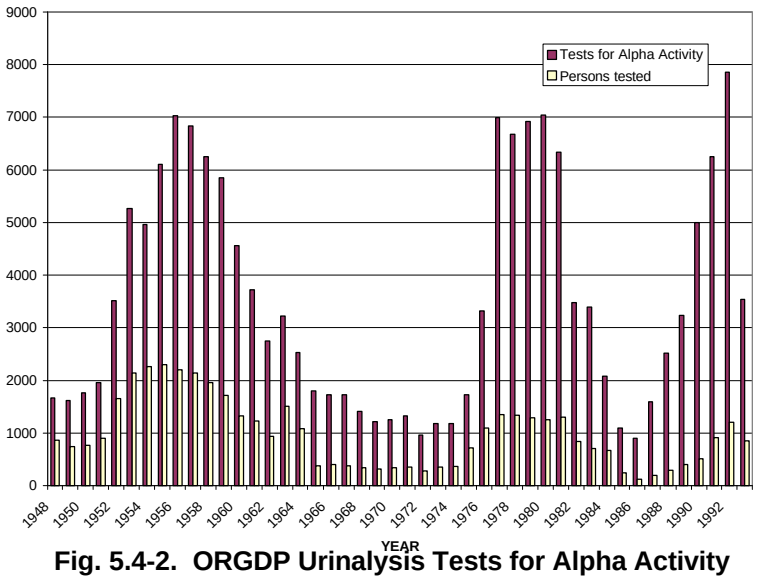

Fig. 5.4-2. ORGDP Urinalysis Tests for Alpha Activity (CY 1948 to FY 1992).

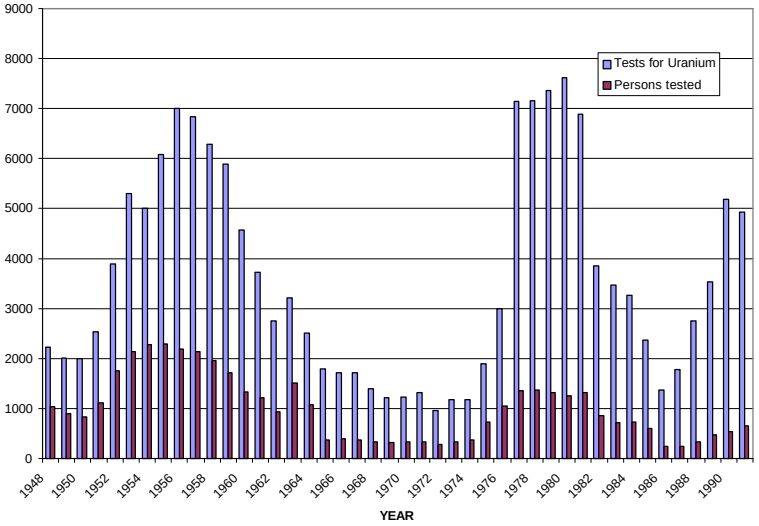

Fig. 5.4-3. ORGDP Urinalysis Tests for Uranium (CY 1948 to FY 1992).

Uranium Analysis

The analytical method for uranium analysis was fluorimetric. Urine samples were prepared and the fluorescence of the sample compared to that of known standards. This analysis was sensitive to the amount of elemental uranium in the sample and did not discriminate between uranium isotopes nor did it detect the presence of transuranics or technetium. Results by department reported in milligrams per liter are shown for CY 1948 through FY 1962 in Figure 5.4-4 and for FY 1963 through FY 1992 in Figure 5.4-5.

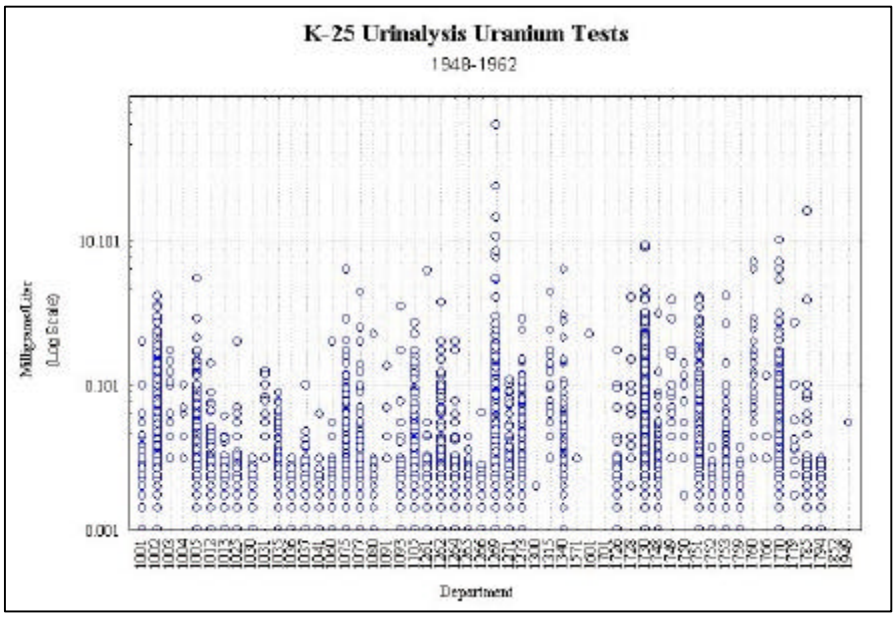

Fig. 5.4-4. Results of Uranium Analyses by ORGDP Department (CY 1948 – FY 1962).

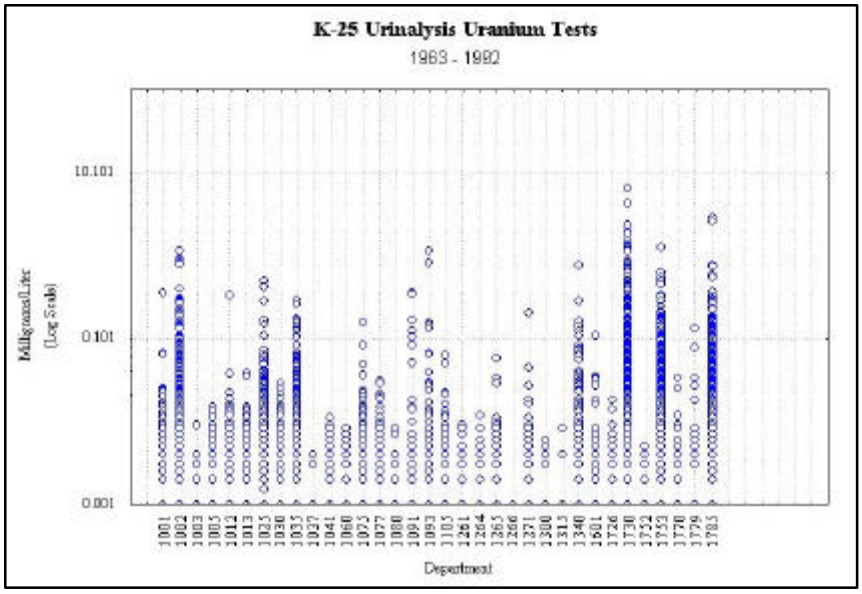

Fig. 5.4-5. Results of Uranium Analyses by ORGDP Department (CY 1963 – FY 1992).

Alpha Activity Analysis

Samples were prepared for alpha counting by evaporating 100 milliliters of urine to dryness with an excess of concentrated nitric acid and igniting the residue over a blast burner. The white salt residue was then dissolved a second time and the uranium present electroplated onto nickel discs. Counting instruments were checked and calibrated against known standards. Background radiation was determined by counting overnight for 480 minutes. Each sample was then counted on two different instruments to assure accuracy. The limit of uncertainty was +/- 1 count per minute. Control samples were counted three times per week. All alpha particles present on the disc were counted including any alpha from the transuranics. Both plutonium and neptunium are alpha emitters with a much greater specific activity (Table 5.4-3) than normal or enriched uranium.

Table 5.4-3. Specific Activity by Material Type.

Material	Specific Activity
Normal Uranium	1.5 dpm per microgram
^{235}U	5 dpm per microgram
Neptunium`	1,550 dpm per microgram
Plutonium	138,000 dpm per microgram

Results by department reported in dpm/100 milliliters are shown for the years CY 1948 through FY 1962 in Fig. 5.4-6 and for years FY 1963 through FY 1992 in Fig. 5.4-7.

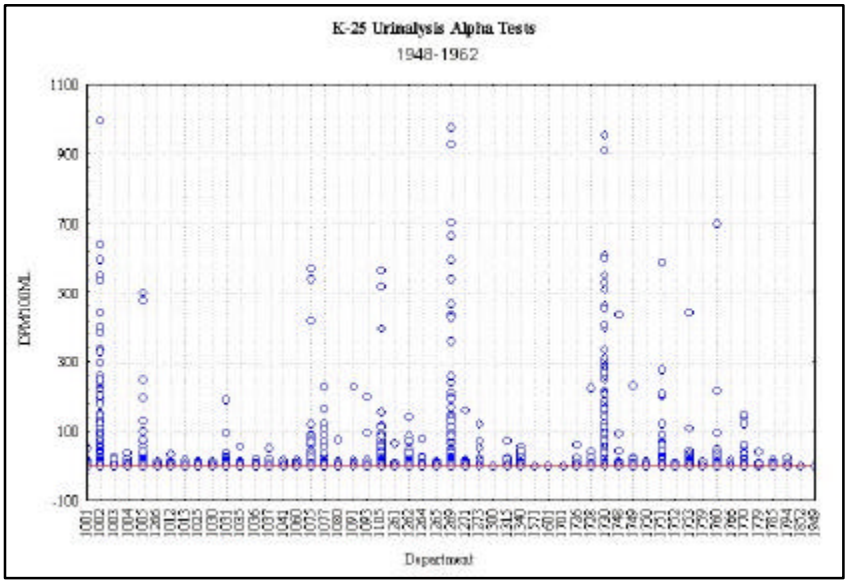

Fig. 5.4-6. Results of Alpha Activity Analyses by Department for CY 1948 to FY 1962.

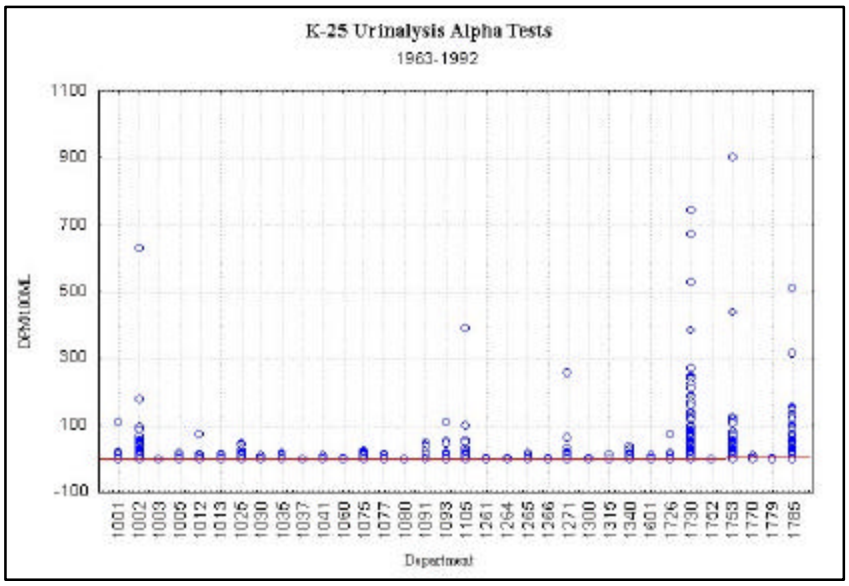

Fig. 5.4-7. Results of Alpha Activity Analyses by Department for FY 1963 to FY 1992.

Comparison of Data

Results from uranium analysis reported in milligrams uranium per liter can be converted to the same units as those reported for alpha activity, dpm per 100 milliliters, if a specific activity for the uranium is estimated. As noted in the table above, the specific activity of natural uranium is 1.5 dpm per microgram. Operations occurring early in the enrichment process such as feed plant activities would most likely be involved with uranium near this natural level of enrichment. Near the product withdrawal points at the top of the cascade the uranium would be enriched to nearly the level of the ^{235}U activity, 5 dpm per microgram. By converting the results of the uranium analysis to dpm per 100 milliliters and comparing to the analysis by alpha activity it can be determined if all the alpha activity present can be attributed to the uranium content of the sample. Alpha activity detected in addition to that of the uranium is contributed by any transuranics present emitting alpha particles, i.e. neptunium or plutonium. The results for the period 1948 to 1962 for two different departments have been sorted in ascending order of activity (dpm/100ml) and compared on Figure 5.4-8 and Figure 5.4-9. Department 1002 (630 employees) was the cascade maintenance department and Department 1035 (104 employees) the respirator service department. In the case of Department 1002 those tests reporting low exposure (about 2,000 analyses) show nominal correlation, that is the level of uranium reported accounts for the alpha activity reported. As the exposure level increases, however the level of alpha activity present is greater than that which could be attributed to uranium, thus implying the presence of transuranics.

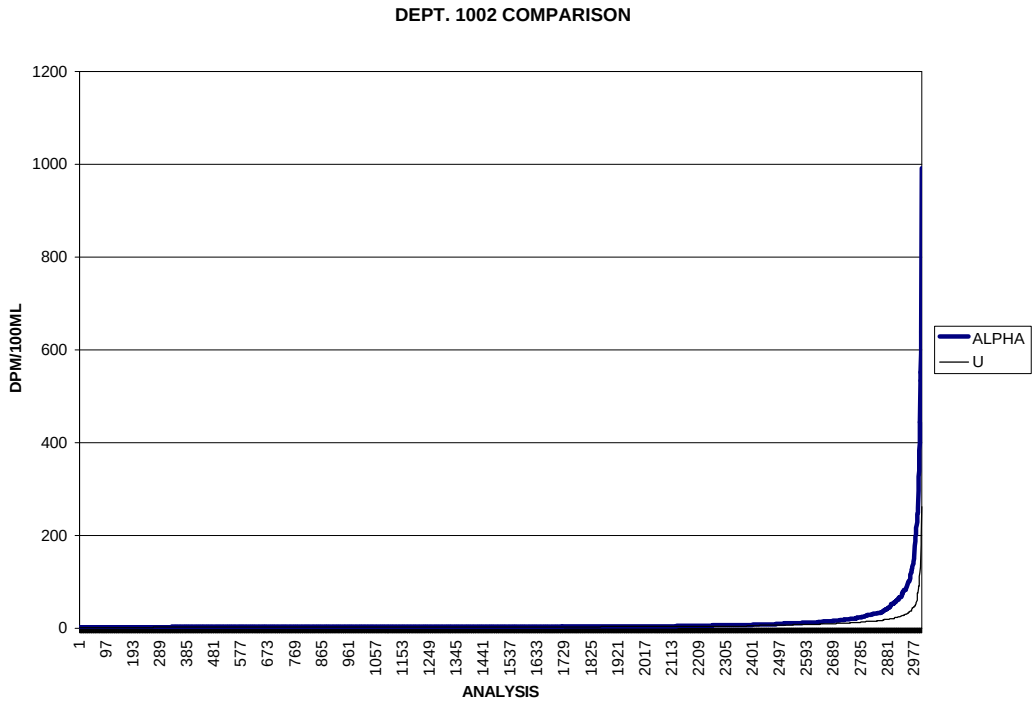

Fig. 5.4-8. Comparison of Uranium and Alpha Activity Results for Department 1002 – Cascade Maintenance.

For Department 1035 this difference is not present. Even as exposure increases the uranium present approximately accounts for the alpha activity reported.

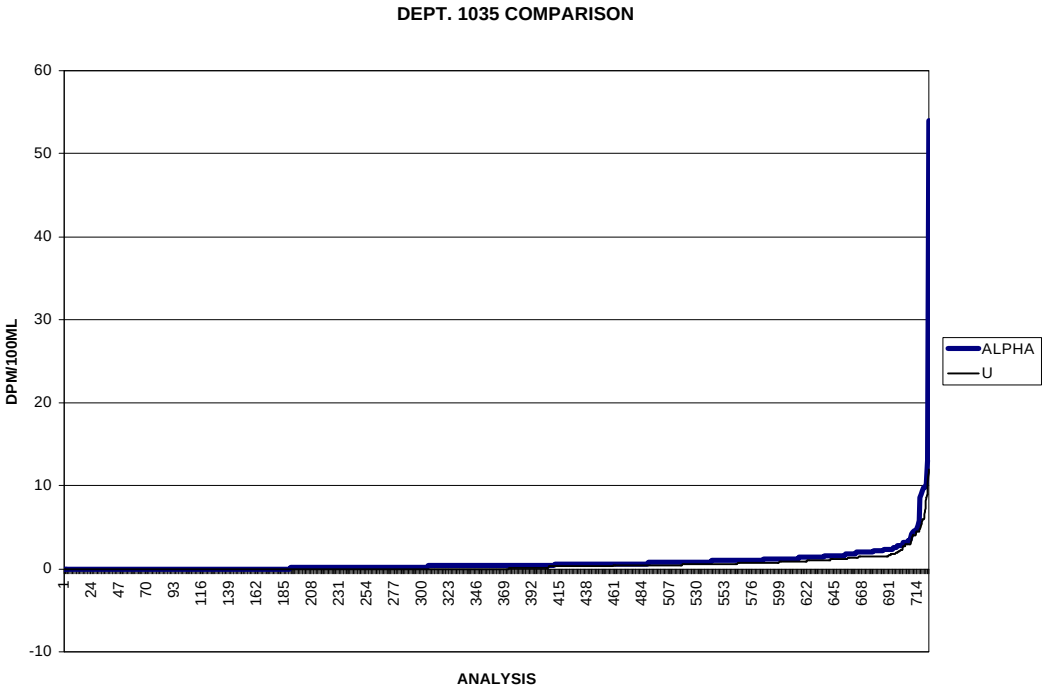

Fig. 5.4-9 Comparison of Uranium and Alpha Activity Results for Department 1035 – Respirator Service.

It should be understood that this comparison of data is of a preliminary nature. A more in-depth assessment will be required to support any conclusions. However, based upon this limited review, it appears that increased levels of alpha activity, due to any significant level of transuranic content in urine, could potentially be determined from the data.

5.4.3 Summary

The potential for worker exposure has been addressed in Table 2.4-1, *Activities Involving Potential Worker Exposure*. This potential was understood, monitored and controlled as evidenced by the use of film badges and the presence of an active urinalysis program for those workers potentially exposed. Further analysis may be warranted to evaluate worker exposure, including detailed dose assessments, as a follow-on to this RU Mass Balance Report.

5.5 POTENTIAL FOR ENVIRONMENTAL CONTAMINATION

5.5.1 Alpha, Beta, and Gamma Activity

General discussions of relative alpha, beta, and gamma activity extracted from the *Quarterly Reports* are shown in Table 5.5-1.

These discussions represent general statements relative to a previous point in time, but they do indicate that a program was in place to monitor activity levels in the plant. Two entries of note in the second and third quarters of 1959 refer to increased air borne contamination resulting from equipment repair and revision operations in K-1131 and K-1231 and to major equipment cleaning in K-1420.

5.5.2 Material Releases

A review of *ORGDP Quarterly Reports*, *Process Engineering Monthly Progress Reports*[14], *Process Engineering Quarterly Progress Reports*[15], and *Production Division Progress Reports*[16] from 1953 through 1966 identified information on material releases. Excerpts from the reports on the material release descriptions, locations, and amounts (where documented) are listed in chronological order in Table 5.5-2.

A brief analysis of the data from June 1958 through December 1965 shows that the kgU and kg^{235}U released per quarter averaged approximately 276 and 1.15, respectively. Several releases were reported in the K-1131 Feed Plant, K-1420, and in various portions of the cascade facilities.

[14] *Process Engineering Progress Report,* Month of May 1958
[15] *Process Engineering Quarterly Progress Reports,* 1959 through 1961
[16] *Production Division Progress Report,* March through May, 1961

Table 5.5-1. Reported Activity Levels Extracted from *ORGDP Quarterly Reports.*

Fiscal Year	Quarter	Alpha	Beta	Gamma
1954	Q1	The alpha contamination level in the plant rose primarily as a result of increased equipment changes, while the penetrating rad levels remained essentially unchanged. One film badge reading of 6 rep represented the highest level ever reported at K-25. A job survey indicated no significant hazard changes had occurred.	A significant decrease occurred in the average beta activity of treated sanitary water while a slight increase in airborne alpha contamination was noted.	
	Q2	Slight increase in alpha contamination attributed to increased maintenance, special tests in certain plant areas and the release of material following a cylinder rupture.	Increase in average beta activity in sanitary water as a result of releases of excessive quantities of fissionable material to the Clinch River.	
	Q3	Reductions in the levels of both penetrating radiation and alpha contamination.		
1955	Q1	Recent increase in alpha surface contamination was noted.		
	Q2		Beta activity in sanitary water increased	
1956	Q1		Beta-Gamma emitting U-daughter products resulted in increase in both rad fields	Beta-Gamma emitting U-daughter products resulted in increase in both rad fields
	Q2		Beta activity in sanitary water supply remained low.	
	Q3		The beta-gamma radiation index increased ~50% as a result of the continued normal accumulation of beta-gamma emitters in the feed production equipment.	A slight increase was noted in the gamma contamination index.
	Q4	Alpha surface contamination throughout the plant remained unchanged.		
1957	Q1	Decreases in airborne alpha activity, penetrating rad index, and the alpha contamination index were noted; A conference on alpha contamination was held for exempt personnel.	Beta activity in sanitary water remained low.	
1958	Q3	Reports occasional high air counts in K-601.		
1959	Q2	Continued increase in plant contamination index, largely associated with remodeling and repair activities in the feed plant and major equipment cleaning in K-1420. Associated increases in airborne alpha emitting materials were noted.		Reference test for correlating gamma survey data with cascade dynamic U-235 inventory.
	Q3	Although alpha contamination decreased ~20%, a marked increase in airborne U concentrations was associated with equipment - revision operations in K-1131 and K-1231 pulverizing building.		

Table 5.5-2. Material Release Information Extracted from ORGDP Reports

Fiscal Year	Quarter	Type Report	Material Release Description	Material Release - Building	Material Release (lbs/UF_6)	Material Release (kg/U)	Material Release ($kg/^{235}U$)
1953	Q2	Plant Quarterly	2506 pounds of UF6 was released accidentally in K-402-1	K-402-1	2,506		
	Q4	Plant Quarterly	Page C-11: Cylinder explosion - 123 pounds of UF6 released to atmosphere - entire building contaminated - K-413, May 25, 1953	K-413	123		
	Q4	Plant Quarterly	June 1, 1953 - Building K-1131 - gland nut valve broke and 952 lbs were released	K-1131	952		
1954	Q1	Plant Quarterly	Five material releases were reported, but were of a minor nature in regard to their effect on working conditions				
1955	Q2	Plant Quarterly	Air activity increases during the calendar year were associate with a number of material releases involving a new product withdrawal area and the feed prep and production facilities				
1956	Q1	Plant Quarterly	Of 8 material releases, 3 involved small amounts of radioactive materials				
1957	Q1	Plant Quarterly	None of the 9 releases of corrosive materials or the 3 releases of U-bearing materials indicated significant problems				
1958	Q4	Engineering Monthly	K-1131 feed plant vent stack losses measured 12 days with normal material and 19 days with 6-range reactor tails material. Vented (kg): Normal 13 kgs U, 0.1 kgs U-235; Reactor Tails 11 kgs U, 0.1 kgs U-235				
1959	Q1	Engineering Quarterly				44.64	0.330
	Q2	Engineering Quarterly				193.00	1.400
	Q3	Engineering Quarterly	5 releases totaling 149,573 grams U and 1.032 grams U-235 (two releases in K-1420 and K-312-1; 2,800 and 20 grams, respectively			150.00	
	Q4	Engineering Quarterly	6 releases of 73,310 grams U, 558 grams U-235			73.00	0.558
1960	Q2	Plant Quarterly				118.00	0.800
	Q3	Plant Quarterly	380,189 gram U and 2,917 grams U-235 released during quarter			380.00	3.000
	Q4	Plant Quarterly	340,908 grams U and 2287 grams U-235 released			341.00	2.000
	Q1	Engineering Quarterly	4 releases of 180,054 grams U, 1,288 grams U-235			180.00	1.300
	Q2	Engineering Quarterly	105,315 grams U, 748 grams U-235			105.00	0.748
	Q3	Engineering Quarterly				132.00	0.900
1961	Q1	Plant Quarterly	185,202 grams U and 1,270 grams U-235			185.00	1.000
	Q2	Plant Quarterly	70,242 grams U as UF6, 651 grams U-235 as UF6			70.00	1.000
	Q3	Plant Quarterly	124,501 grams U, 996 grams U-235			125.00	1.000
	Q1	Engineering Quarterly	Table showing where releases were shows most came from K-1131 vent stacks	K-1131		148.00	1.000
	Q2	Engineering Quarterly	134,774 grams U, 957 grams U-235			135.00	0.957
	Q3	Engineering Quarterly	9 releases totaling 128,693 grams U, 1,028 grams U-235			128.69	1.028
	Q4	Production Div Quarterly				288.00	2.000
1962	Q1	Plant Quarterly	184,196 grams U, 1,475 grams U-235			185.00	1.000
	Q2	Plant Quarterly	212,769 grams U, 622 grams U-235			213.00	1.000
	Q3	Plant Quarterly	6,774 grams U, 2,609 grams U-235			7.00	3.000
	Q4	Plant Quarterly	348,085 grams U, 2,526 grams U-235 as UF6			348.00	3.000
	Q4	Plant Quarterly	3,955 grams U, 395 grams U-235			4.00	0.400
1963	Q1	Plant Quarterly	6,689 grams U, 91 grams U-235			7.00	
	Q3	Plant Quarterly	67 grams U, 14 grams U-235	K-1420			
	Q4	Plant Quarterly	Table G-1 shows 496 grams of uranium released				
1964	Q3	Plant Quarterly	4 material releases (see Table G-1) accounted for the release of 3,862 grams U and 95 grams U-235 to the atmosphere as UF6			3,862.00	
	Q4	Plant Quarterly	2 uranium releases - 22.6 kgsU and 0.068 kgs U-235 were vented to the atmosphere from K-902-1, cell 8	K-902-1		22.60	0.068
1965	Q2	Plant Quarterly				0.61	0.001
1966	Q2	Plant Quarterly	5,213 grams U, 194 grams U-235			5.21	0.194

ORGDP Material Release Reports from 1957 through 1961[17] document the grams of uranium released during this time period. These data are presented graphically in Fig.5.5-1.

[17] *ORGDP Material Release Reports*, 1957 through 1961

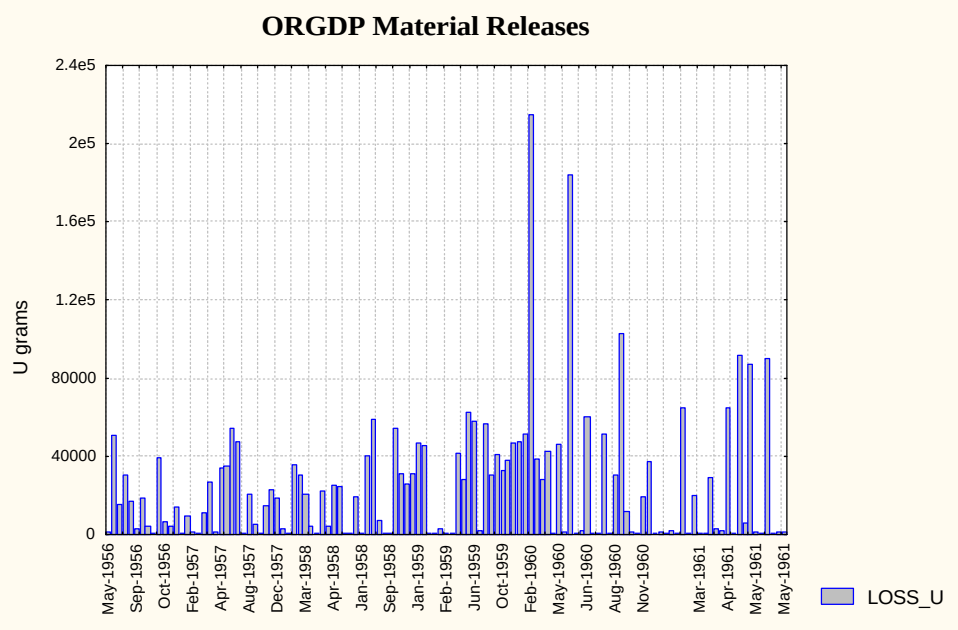

Fig. 5.5-1. ORGDP Material Releases.

5.5.3 Environmental Monitoring

As presented in Section 2.5, environmental monitoring of the air and water adjacent to the Oak Ridge Reservation has been an on-going program since the 1950s. The most exhaustive compilation of environmental data resulted from the Oak Ridge Dose Reconstruction Project, documented in a series of task reports in July 1999. The Task 6 report[18] entitled *Uranium Releases from the Oak Ridge Reservation – a Review of the Quality of Historical Effluent Monitoring Data and a Screening Evaluation of Potential Off-Site Exposures draws the following conclusion:*

> "…the K-25/S-50 uranium releases are candidates for further study, but (they) are not high priority candidates for further study."

The Task 7 report[19] entitled *Screening-Level Evaluation of Additional Potential Materials of Concern* performed qualitative and quantitative screening of various materials of concern at ORGDP (and the other ORR sites), including ^{237}Np and ^{99}Tc. Based on the analysis of data, it was determined that Np did not warrant further study. Technetium was identified as one of the potential candidates for further study, but was not determined to be a high priority.

[18] *Uranium Releases from the Oak Ridge Reservation – a Review of the Quality of Historical Effluent Monitoring Data and a Screening Evaluation of Potential Off-Site Exposures*, PD-02314; Reports of the Oak Ridge Dose Reconstruction, Vol. 5, The Report of Project Task 6, July 1999.
[19] *Screening – Level Evaluation of Additional Potential Materials of Concern*, PD-02315; Reports of the Oak Ridge Dose Reconstruction, Vol. 6, The Report of Project Task 7, July 1999.